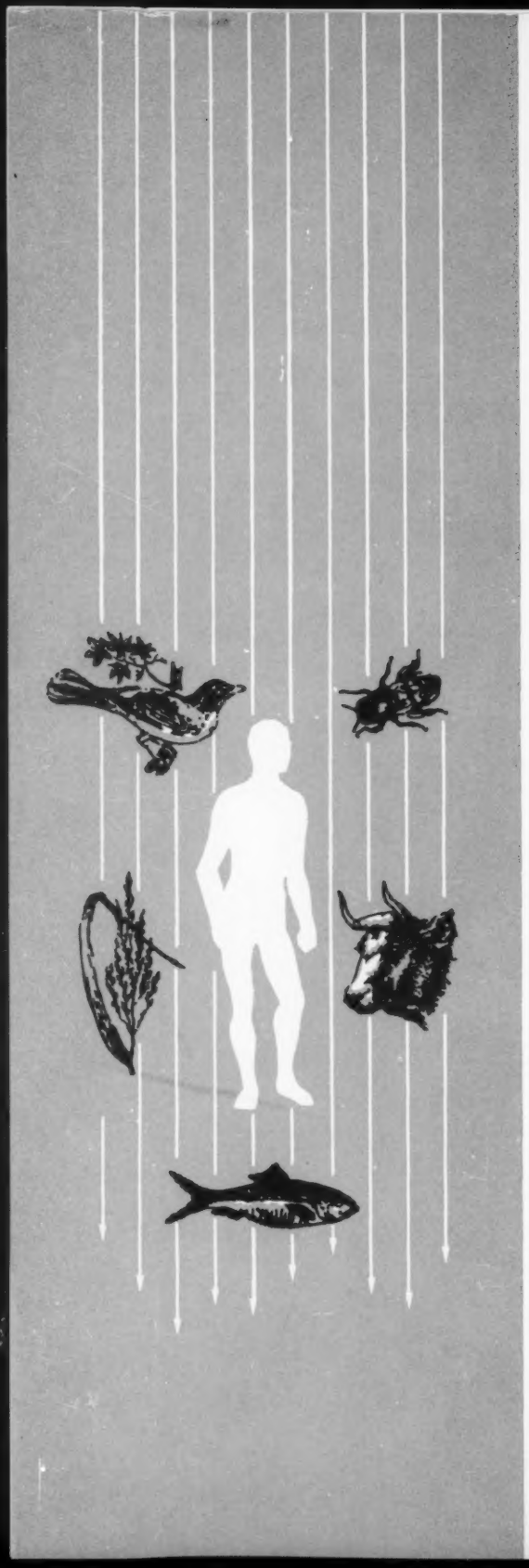

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The Working Group is comprised of representatives of the U.S. Departments of Agriculture; Commerce; Defense; the Interior; Health, Education, and Welfare; State; Transportation; and Labor; and the Environmental Protection Agency.

The Monitoring Panel consists of representatives of the Agricultural Research Service, Animal and Plant Health Inspection Service, Extension Service, Forest Service, Department of Defense, Fish and Wildlife Service, Geological Survey, Food and Drug Administration, Environmental Protection Agency, National Marine Fisheries Service, National Science Foundation, and Tennessee Valley Authority.

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Pesticide monitoring activities of the Federal Government, particularly in those agencies represented on the Monitoring Panel which participate in operation of the national pesticides monitoring network, are expected to be the principal sources of data and articles. However, pertinent data in summarized form, together with discussions, are invited from both Federal and non-Federal sources, including those associated with State and community monitoring programs, universities, hospitals, and nongovernmental research institutions, both domestic and foreign. Results of studies in which monitoring data play a major or minor role or serve as support for research investigation also are welcome; however, the Journal is not intended as a primary medium for the publication of basic research. Publication of scientific data, general information, trade names, and commercial sources in the *Pesticides Monitoring Journal* does not represent endorsement by any Federal agency.

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CONTENTS

Volume 15 June 1981 Number 1

Page

FISH, WILDLIFE, AND ESTUARIES

Polychlorinated Biphenyls and Other Organic Chemical Residues in Fish from Major United States Watersheds Near the Great Lakes, 1978 ____ 1
Gilman D. Veith, Douglas W. Kuehl, Edward N. Leonard, Kenneth Welch, and Glenn Pratt

Organochlorine Pesticide Residues in Some Indian Wild Birds ____ 9
Bhupendra S. Kaphalia, Mirza M. Husain, Tejeshwar D. Seth, Ashwini Kumar, and Coimbatore R. Krishna Murti

Cadmium, Lead, Mercury, Arsenic, and Selenium Concentrations in Freshwater Fish, 1976–77—National Pesticide Monitoring Program ____ 14
Thomas W. May and Gerald L. McKinney

FOOD AND FEED

Pesticide, Heavy Metal, and Other Chemical Residues in Infant and Toddler Total Diet Samples—(II)—August 1975–July 1976 ____ 39
Roger D. Johnson, Dennis D. Manske, Dallas H. New, and David S. Podrebarac

Organochlorine Pesticides and PCBs in Cod-Liver Oil of Baltic Origin, 1971–80 ____ 51
Jerzy Falandysz

Pesticide, Metal, and Other Chemical Residues in Adult Total Diet Samples—(XII) —August 1975–July 1976 ____ 54
Roger D. Johnson, Dennis D. Manske, and David S. Podrebarac

APPENDIX ____ 70

ERRATUM ____ 72

MEETING NOTICE ____ 73

Information for Contributors ____ 74

FISH, WILDLIFE, AND ESTUARIES

Polychlorinated Biphenyls and Other Organic Chemical Residues in Fish from Major United States Watersheds Near the Great Lakes, 1978

Gilman D. Veith,[1] Douglas W. Kuehl,[2] Edward N. Leonard,[1] Kenneth Welch,[2] and Glenn Pratt[3]

ABSTRACT

Twenty-six composite samples of fish were collected during 1978 from United States watersheds near the Great Lakes and analyzed for polychlorinated biphenyls (PCBs) and related organic chemicals. PCB mixtures resembling Aroclor 1254 were found in all samples, and mixtures resembling Aroclor 1242 (or 1016) were found in 77 percent of the samples. Total PCB concentrations in the whole-fish composite samples ranged from 0.13 to 14.6 ppm; 65 percent of the samples contained > 2 ppm PCBs. DDT and its metabolites were also found in all samples. ΣDDT concentration was 1.66 ppm, and 81 percent of the samples contained < 1.0 ppm ΣDDT. Chlordane ranged from < 0.001 to 2.57 ppm in 38 percent of the samples. Hexachlorobenzene was found in 65 percent of the samples, ranging from <0.005 to 0.447 ppm. Other chemicals identified by gas chromatography/mass spectrometry included petroleum hydrocarbons and chlorobenzenes, chlorostyrenes, chlorophenols, and chlorinated aliphatic compounds. Fish from the Ashtabula River (Ohio), Rocky River (Ohio), and Wabash River (Indiana) contained extremely complex residues of chlorinated and other organic chemicals.

Introduction

In 1976, authors extended their gas-liquid chromatography/mass spectrometry (GLC/MS) exploratory studies of organic chemical residues in Great Lakes fish to include residues in fish from major United States rivers for the purpose of tabulating polychlorinated biphenyls (PCBs) and other xenobiotic chemicals accumulating in the aquatic environment. A previous work (*6*) showed that the types and concentrations of chemical residues in fish varied immensely among rivers in the same area of the country. Fish in some rivers in eastern Michigan and Ohio contain PCB residues almost exclusively, whereas fish from the Ashtabula River nearby in Ohio contain at least 19 major chlorinated chemicals in addition to PCBs. Fish from rivers a few miles apart differ in hexachlorobenzene (HCB) residues by a factor of almost 3,000.

The wide variation in both the types and amounts of chemicals in different waters suggests that it is not cost-effective to apply trend-monitoring programs to the problem of determining the extent of contamination by toxic chemicals. Trend monitoring requires a predetermined list of chemicals to monitor, precise methods for measuring small differences in concentration, and, to minimize biological variability, a fairly rigid sampling protocol with respect to species and size. In contrast, the initial problem regarding toxic industrial chemicals is the identification of every major chemical component of residues from hundreds of industrial areas. Such areas may not have diverse fish populations because of the contamination. Finally, order-of-magnitude estimates of residue concentrations by GLC/MS may be used to direct enforcement-related field studies to "hot-spots" for more intensive studies.

A previous work (*6*) indicated that taking composite samples of any fish near the mouth of a river provides a convenient, enriched sample of bioaccumulable chemicals being discharged into the entire watershed and excludes the bulk of the less persistent, nonaccumulable chemicals attributable to natural products and sanitary wastes. Because the accumulation of chemicals in fish species varies considerably less than the concentration of chemicals in rivers and areas of rivers, composite samples also provide adequate estimates of relative concentrations in the sampling areas. This paper presents the results of exploratory studies of chemicals in fish from rivers in Minnesota, Wisconsin, Indiana, Michigan, Ohio, and New York (Figure 1).

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FIGURE 1. *Map of U.S. EPA Region V showing sites where fish samples were collected for GLC/MS analysis of bioaccumulating hazardous organic chemical residues*

Materials and Methods

COLLECTION OF FISH

The areas that were sampled for detailed GLC/MS analyses in 1976 and the 26 sampling areas for the present study (1978) are shown in Figure 1. Table 1 gives a brief description of the 1978 sampling locations.

State and federal field personnel used nets and other conventional apparatus to collect fish. Sampling areas included known problem areas disclosed by previous studies as well as rivers for which little data were available. Where possible, samples were collected near mouths of rivers or confluence of major tributaries. Sample areas are designated as general areas of rivers rather than exact locations because of the migratory nature of many fish and the tendency of bioaccumulable chemicals to contaminate large areas near the discharge. Samples were wrapped in solvent-rinsed aluminum foil, frozen, and shipped with dry ice to the Environmental Research Laboratory in Duluth, Minnesota.

PREPARATION OF SAMPLES

The procedures for preparation and analysis of samples have been described previously (*6*). Composite whole-fish samples were homogenized with a Hobart tissue grinder. Subsamples weighing 20 g were extracted in a Soxhlet extractor with a 1:1 mixture of hexane and methylene chloride. Following Florisil column cleanup, samples were analyzed by electron-capture (EC) and flame-ionization (FI) GLC and multiple-ion-detection (MID) GLC/MS.

GLC analysis provided measurement of PCBs, HCB, DDE, and ΣDDT, as well as information on the complexity of the residue with respect to other nonpolar organic chemicals. MID GLC/MS was used to quantitate *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, heptachlor epoxide, and oxychlordane. A procedural blank was performed with every fifth sample. Duplicate analyses of several samples gave results within 4 percent for both EC GLC and MID GLC/MS. Recovery from laboratory-raised fathead minnows spiked with 1 ppm PCB and 50 ppb pesticides was >92 percent for each compound.

A second subsample (100 g) of each fish was Soxhlet-extracted in a 1:1 mixture of hexane and methylene chloride, cleaned by gel-permeation chromatography (*2*), and qualitatively analyzed by full mass range

TABLE 1. *Concentrations of major chlorinated hydrocarbons in composite whole-fish samples from 22 United States watersheds near the Great Lakes, 1978*

SAMPLING AREA	SAMPLE COMPOSITION	PCB RESIDUES, PPM			DDE, PPM	ΣDDT, PPM	HCB, PPB	CHLORDANE (CIS + TRANS), PPB	NONACHLOR (CIS + TRANS), PPB	HEPTACHLOR, PPB	HEPTACHLOR EPOXIDE, PPB	OXYCHLORDANE, PPB
		1016/1242	1254	TOTAL								
Wisconsin River, below Nekoosa Dam, Wisc.	5 walleye 2 carp 2 white sucker	0.38	0.86	1.24	0.08	0.14	7	*	*	*	*	*
Fox River, below DePere Dam, Wisc.	4 carp 4 white sucker 5 channel catfish	3.61	6.41	10.0	0.75	1.06	18	*	17	*	*	*
Lake Pepin, Mississippi River, Minn.	3 carp	0.15	4.35	4.50	0.36	1.03	37	*	20	*	*	*
Wabash River, below Terre Haute, Ind.	5 channel catfish	0.31	1.77	2.08	0.08	0.15	<5	45	37	27	5	*
Wabash River, above Terre Haute, Ind.	5 carp	0.18	1.07	1.25	0.13	0.21	<5	21	25	*	*	*
White River, Paragon, Ind.	11 channel catfish	0.51	2.65	3.16	0.15	0.47	7	239	288	*	*	*
St. Joseph River, Berrien Springs Dam, Mich.	4 carp	0.34	2.62	2.96	0.35	0.76	9	*	24	*	*	*
Grand River, Clinton County, Mich.	3 carp	<0.10	4.91	4.91	0.56	0.31	36	2,570	3,070	*	*	*
Tittabawassee River, Smith's Crossing, Mich.	5 channel catfish 2 carp	1.25	1.40	2.65	0.23	0.36	64	*	4	*	*	*
Raisin River, Monroe County, Mich.	4 carp	9.83	4.80	14.6	0.25	0.48	14	*	36	*	*	*
Scioto River, Rosemont, Ohio	1 freshwater drum 2 carp 1 channel catfish 1 quillback	0.32	1.98	2.30	0.12	0.40	7	64	125	*	*	*
Ashtabula River, Ashtabula, Ohio	1 white sucker 1 hog sucker 2 stone rollers 1 carp	9.36	1.52	10.9	0.79	0.92	447	*	*	77	*	*
Huron River, Ohio	1 goldfish 2 freshwater drum 2 carp	<0.10	0.62	0.62	0.17	0.26	<5	*	*	8	*	*
Chagrin River, Ohio	3 freshwater drum 2 carp	0.26	3.57	3.83	0.50	0.82	<5	51	434	*	*	*
Rocky River, Ohio	2 carp 1 goldfish	0.84	4.54	5.38	0.52	0.88	21	2,680	1,820	*	*	167
Conneaut River, Ohio	4 freshwater drum 1 carp	<0.10	0.40	0.40	0.04	0.09	39	*	*	*	*	*
Black River, Ohio	5 channel catfish 1 freshwater drum	0.16	1.62	1.78	0.10	0.31	<5	*	*	*	*	*
Portage River, Ohio	5 carp	0.35	2.66	3.01	0.46	0.72	<5	*	*	*	*	*
Sandusky River, Ohio	5 freshwater drum	<0.10	0.59	0.59	0.08	0.15	<5	*	*	*	*	*
Maumee River, Waterville, Ohio	5 carp	<0.10	0.25	0.25	0.02	0.05	<5	25	25	*	*	*
Maumee River, Collen Park, Ohio	3 carp	1.38	3.38	4.76	0.23	0.53	24	*	*	*	*	*
Great Miami River, Miamisburg, Ohio	2 carp 3 goldfish	2.00	5.18	7.18	0.24	0.45	13	24	16	11	*	*
Great Miami River, Elizabethtown, Ohio	3 carp 1 channel catfish	0.78	8.59	9.37	0.66	1.32	13	43	25	*	*	*
Lake Ontario, near Oswego, N.Y.	2 brown trout	1.79	3.97	5.76	1.00	1.66	87	*	*	2	*	1
Cattaragaus Creek, N.Y.	5 white sucker 2 hog sucker 1 brown trout	<0.10	0.13	0.13	0.01	0.03	<5	*	*	*	*	*
Cattaragaus Creek Mouth, N.Y.	3 coho salmon	0.16	1.59	1.75	0.21	0.40	7	*	*	9	*	3

NOTE: * = Below the detection limit of 0.5 ppb.

(m/z 50–550) scanning GLC/MS. Again, procedural blanks were performed with every fifth sample, and spiking experiments showed >90 percent recovery.

ANALYSES

The GLC/MS analyses were performed on a Finnigan 4000 mass spectrometer interfaced to an INCOS data system. Instrument parameters and operating conditions follow:

Column:	glass, capillary, 30-m long by 0.25-mm (ID), coated with SE-30
Temperature, °C:	column programmed from 100 to 225° at 4°/minute (with a 20-minute hold) ion source 280
Carrier gas:	helium flowing at 30 cm/second
Mass scan:	50–550/2 seconds
Electron energy:	70 eV
Emission:	350 mA

MID GLC/MS analyses were also performed on the above equipment, with the same temperature programming, followed by a 20-minute hold. The mass spectrometer was computer-controlled to monitor six ions for equal time during a 2-second period. The six ions were m/z 373 for chlordane, m/z 409 for nonachlor, m/z 272 for heptachlor, m/z 355 for heptachlor epoxide, m/z 387 for oxychlordane, and m/z 442 for defluorotriphenylphosphene (DFTPP).

DFTPP was used as an internal standard for MID quantification. Each extract was spiked with DFTPP to give a concentration of 10 ng/μl. Two standards of the chlordane components of 5 ng/μl and 20 ng/μl each were used as samples after each set of five fish samples. Quantification was based on a 10-ng/μl solution with standard INCOS software. The quantified standards gave values within 4 percent of expected values. The limit of detectability was 0.50 ppb wet weight.

Ion source was operated as above with an emission of 350 mA.

Results and Discussion

PCBs were found in all 26 samples at concentrations ranging from <0.1 ppm in fish from the upper reaches of Cattaragaus Creek, New York, to 14.6 ppm in fish from the Raisin River, Michigan. Aroclor 1254 constituted over 50 percent of the total PCB residues in the majority of the samples. Aroclor 1016/1242 was found in 20 of the 26 samples at concentrations ranging from <0.1 to 9.83 ppm in Raisin River fish. These results are consistent with the authors' previous study of 58 samples, in which Aroclor 1016/1242 was present in 77 percent of the samples (*6*).

Fish from the Fox River (Wisconsin), Raisin River (Michigan), and Ashtabula and Greater Miami Rivers (Ohio) remain heavily contaminated with PCBs. Twelve other samples contained PCB residues >2 ppm. Although PCB concentration in the edible portion of the fish is expected to be lower than that in the whole fish, the present data suggest that 65 percent of the fish samples would pose significant hazards to animals, such as mink, feeding on the fish (*1*).

DDT, once the major organochlorine contaminant in fish in many U.S. waterways, is a minor contaminant in the river systems investigated in the present study. Although DDE was found in all samples, ΣDDT concentration in 81 percent of the samples was below 1.0 ppm; the maximum concentration was 1.66 ppm, in Lake Ontario fish.

HCB was the next most prevalent organochlorine found in the present study; 65 percent of the samples contained measurable quantities. Although most samples contained <0.05 ppm HCB, Ashtabula River fish contained 0.447 ppm. The authors' previous work (*6*) revealed concentrations of 3.14 ppm HCB in fish from the Ashtabula River in 1976. The apparent decline may be the result of sampling variability or of pollution-abatement measures taken since the 1976 discovery. The fact that fish analyzed in the present study were collected upstream from the alleged discharge (*3*) into the Ashtabula River may also be significant. An ongoing field investigation is a direct result and a primary benefit of this type of biomonitoring, because areas of highest contamination are identified for more intensive study at minimum cost.

Chlordane and components of technical chlordane were found in 38 percent of the samples. Although fish from most of the rivers contained <0.05 ppm chlordane, fish from the Grand River, Michigan, and Rocky River, Ohio, contained 2.57 ppm and 2.68 ppm, respectively. The total nonachlor concentrations in these fish were 3.07 ppm and 1.82 ppm, respectively. Heptachlor and heptachlor epoxide were found only in fish from the Wabash, Ashtabula, and Huron Rivers and Lake Ontario. Oxychlordane was present in Rocky River, Lake Ontario, and Lake Erie fish at a maximum concentration of 167 ppb. The coho salmon caught at the mouth of the Cattaragaus Creek, New York, are undoubtedly Lake Erie fish, which would account for the oxychlordane residues.

In addition to chemicals quantified by GLC or MID GLC/MS, the results of exhaustive GLC/MS studies of the fish extracts are summarized in Table 2. The first 25 organic chemicals identified in Table 2 are aliphatic and aromatic hydrocarbons. Heptadecane, pentadecane, and related hydrocarbons are natural products of benthic algae, as well as the results of petroleum contamination from mixtures such as fuel oil. Present methodology permits only qualitative statements about the sources of these compounds, based on FI GLC chromatograms.

Pyridinecarboxamide appeared to be as common as heptadecane in fish from these rivers.

Pentachloroanisole was identified in 15 of the 26 samples of fish. The authors have observed halogenated anisoles in effluents of sewage treatment plants receiving the respective halogenated phenols (*4*). Present information suggests that the anisoles arise from methylation of the corresponding phenols by bacteria. Studies at the Environmental Research Laboratory in Duluth (unpublished data) have shown that fish exposed to halogenated phenols do not produce the anisoles metabolically. The presence of pentachloroanisole may therefore arise from the widespread use of pentachlorophenol as a wood preservative. Because the bioconcentration factor for the methyl derivative of phenols is several orders of magnitude greater than that of the phenol, pentachloroanisole as an environmental contaminant is probably the result of selective bioaccumulation of a more persistent metabolite of pentachlorophenol.

Heptachloronorbornene and hexachloronorbornadiene were found only in the Wabash River fish collected below Terre Haute, Indiana. These two compounds are intermediate chemicals in the production of cyclodiene pesticides, and their occurrence is linked to manufacturing sites for these pesticides. These chemicals were included in Table 2 because they are unique in the Wabash River, not because of widespread occurrence. These data confirm the authors' studies of fish from the lower Wabash (*6*) and suggest a source of contamination in the vicinity of Terre Haute, Indiana. The only other identification of heptachloronorbornene and hexachloronorbornadiene in the aquatic environment was reported by the Food and Drug Administration, U.S. Department of Health and Human Services, in fish from the Mississippi River below Memphis, Tennessee (*5*).

Mirex was identified only in Lake Ontario fish. The compound was first observed in 1973 and was extensively investigated by state and federal agencies.

PCBs were identified in all samples and, although the tetra-, penta-, and hexachlorobiphenyl homologs were predominant, PCBs containing two or three chlorine atoms were found in 19 of the 26 samples.

The above-mentioned quantitative and qualitative data present a reasonably comprehensive description of the chemical residues that contaminate fish in some major rivers. However, these data fail to illustrate adequately the need for improved biomonitoring or analytical methods development. Fish from many of the rivers contain residue mixtures that are similar to the GLC chromatogram of the Maumee River fish presented in Figure 2. This chromatogram was obtained by using a 30-m, wall-coated capillary column and an electron-capture detector. Although there are many chemicals in this extract, PCBs, HCB, and natural products account for all peaks in the chromatogram. Therefore, the residues in this area are comparatively simple to work with, and routine GLC methods should be adequate for any surveillance work.

In contrast, the Ashtabula, Wabash, and Tittabawassee Rivers contain extremely complex mixtures of bioaccumulable chemicals. An electron-capture capillary chromatogram of extract of fish from the Ashtabula River is shown in Figure 3. Chemicals identified include tetra-, penta-, and hexachlorobutadiene; chlorinated benzenes up to hexachlorobenzene; penta- and hexachlorobutylamines; and numerous hexa-, hepta-, and octachlorostyrenes. Despite the identification of almost 100 chemicals in this sample, those identified to date are largely only those in the highest concentrations. A comprehensive study of residues in fish from the Ashtabula and Wabash Rivers is presented elsewhere (*3*).

Even though progress has been made in developing methods for rapid characterization of chemical residues, the long lists of chemicals being published from studies of environmental samples should not lead to the conclusion that present methods are thorough or adequate. In many of the rivers the authors have studied during the past four years, chemicals are present that cannot be identified without improved cleanup methods and instrumental techniques. More important, the number of sample sites studied would have to be increased by an order of magnitude in order to screen even a single sample from industrial areas over the next five years. Major improvements in the current state-of-the-art methods for GLC/MS screening will have to be made before an adequate number of samples can be thoroughly studied.

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TABLE 2. *Chemical contaminants identified by GLC/MS in fish from United States rivers near the Great Lakes, 1978*

Chemical Contaminant	Wisconsin River	Fox River	Lake Pepin	Wabash River (above Terre Haute)	Wabash River (below Terre Haute)	White River	St. Joseph River	Grand River	Tittabawassee River	Raisin River	Scioto River	Ashtabula River
Tridecane	1			2						2		
Tetradecane				2						2		
Pentadecane							2					
Hexadecene			1									
Hexadecane	1		1	1								
Heptadecene							2	2				
Heptadecane				1			1	1	1	1	1	
Octadecane				1							1	
Nonadecane								1				
Eicosane								2				
Naphthalene					1		1			1		
Methylnaphthalene					2	2	2	2		2		
Dimethylnaphthalene					2		2	2		2		
Biphenyl						1						
Methylbiphenyl						2		2		2		
C2-Biphenyl						2		2				
Phenanthrene				2	2	2	2	2		2		
Fluoranthene				2	2	2	2	2		2		
Pyrene					2	2	2	2		2		
Fluorene												
Dibenzofuran												
Acenaphthalene												
Methylbenzanthracene												
Dibenzothiophene												
Pyridinecarboxamide	1	1			1			1	1	1	1	
Terphenyl												
Phenylnaphthalene												
Pentachlorobenzene									1		1	1
Hexachlorobenzene	1	1	1			1	1	1	1	1	1	1
Pentachloroanisole						1	1	1			1	1
Pentachlorophenol					1							
Pentachlorobenzyl alcohol					2							
DDE			1		1	1	1	1	1	1		
TDE			1			1	1	1	1	1		
DDMU			1		1	1	1					
DDT			1		1	1	1	1	1	1		
Heptachloronorbornene					2							
Heptachloronorbornadiene					2							
Mirex												
Photomirex												
Endrin					1							
Monochlorobiphenyl										2		
Dichlorobiphenyl	2									2		
Trichlorobiphenyl	2	2	2		2	2	2	2	2	2		
Tetrachlorobiphenyl	2	2	2	2	2	2	2	2	2	2	2	2
Pentachlorobiphenyl	2	2	2	2	2	2	2	2	2	2	2	2
Hexachlorobiphenyl			2	2	2		2	2	2	2	2	2
Heptachlorobiphenyl			2	2	2	2	2	2	2	2	2	2
Octachlorobiphenyl							2	2	2			
Hexachlorostyrene												2
Heptachlorostyrene												2
Octachlorostyrene												1
Chlordane				1	1	1		1			1	
Nonachlor		1	1	1	1	1	1	1	1	1	1	
Heptachlor					1							1
Heptachlor epoxide					1							
Oxychlordane												

NOTE: 1 = Confirmed by GLC and MS retention time data; 2 = confirmed by MS data; 3 = suggested by MS data.

TABLE 2 (cont'd.).

Huron River	Chagrin River	Rocky River	Conneaut River	Black River	Portage River	Sandusky River	Maumee River (Waterville)	Maumee River (Collen)	Great Miami River (Miamisburg)	Great Miami River (Elizabethtown)	Lake Ontario	Cattaragaus Creek	Cattaragaus Creek (Mouth)
		2			2								
		2			2								
											2		
	2	1	1							1	1	1	1
1	1	1	1		1	1	1			1			
1			1							1			
1			1				1			1			
			2							2			
		2	2	2									
		2	2										
		1		1									
								2		2			
		2	2	2						2			
2		2	2	2	2				2				2
2		2	2	2					2	2			
2		2	2	2					2	2			
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			2		2								
				2									
								2					
		2		2									
1	1		1	1	1	1	1	1	1	1	1	1	1
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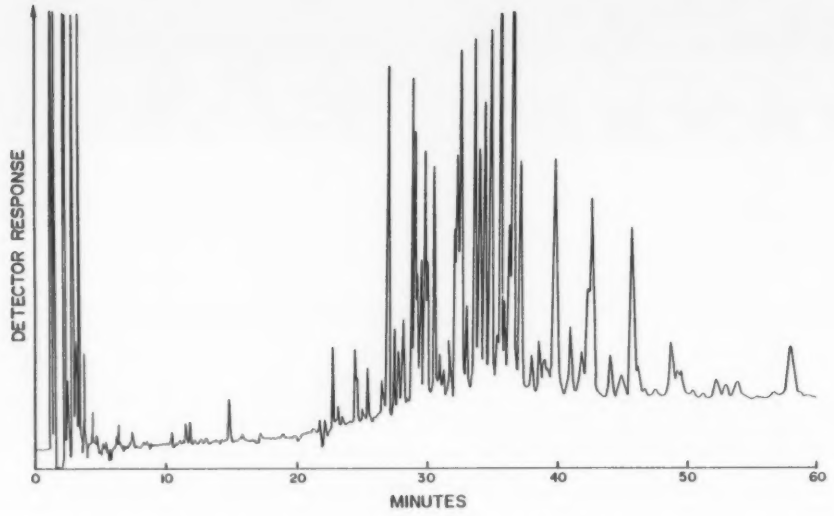

FIGURE 2. *Capillary electron-capture chromatogram of fish extract from Maumee River, Ohio.*

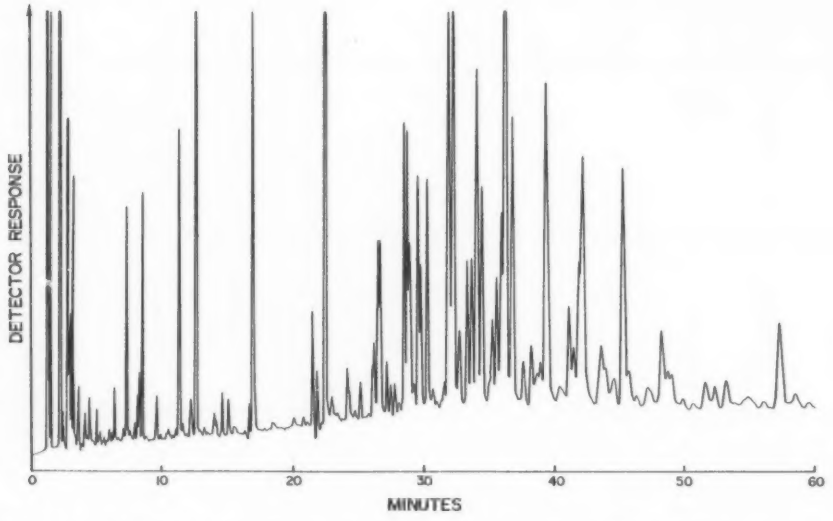

FIGURE 3. *Capillary electron-capture chromatogram of fish extract from Ashtabula River, Ohio.*

Organochlorine Pesticide Residues in Some Indian Wild Birds [1]

Bhupendra S. Kaphalia, Mirza M. Husain, Tejeshwar D. Seth, Ashwini Kumar, and Coimbatore R. Krishna Murti

ABSTRACT

Residues of BHC and DDT were estimated by gas-liquid chromatographic analysis of the internal body organs, depot fat, and blood plasma of a few species of Indian wild birds captured in and around the urban area of Lucknow. Total BHC and γ-BHC (lindane) levels were high in breast muscle, liver, heart, and lung tissues of pigeon, crow, and vulture, compared with the respective tissues of chicken, cattle egret, and kite. More lindane and total BHC was found in tissues of vulture compared with other species. The major part of BHC isomers in the brain of birds examined was accounted for by α-BHC. Total BHC detected in depot fat of crows was 29.7 ppm; lesser amounts were found in vulture, kite, and cattle egret, respectively. Total DDT levels were comparable in the blood plasma of chicken, pigeon, crow, and cattle egret, although residues generally showed the following order in the tissues examined: chicken < pigeon < cattle egret < crow < kite < vulture. High levels of DDT were detected in depot fat of crow, kite, and vulture (50.8, 67.0, and 95.3 ppm, respectively). Avian species thus reflect biological magnification of BHC and DDT residues, presumably due to their food habits.

Introduction

Organochlorine pesticides and related compounds have been detected in significant amounts in the environment and in human body tissues (*8*). Pesticides are dispersed both during their manufacture and by their extensive use for controlling vector-borne diseases and crop pests.

Environmental contamination from persistent organochlorines has been recognized as a threat to wildlife for more than two decades. Many residues have been found in tissues and eggs of birds in Europe and North America (*7, 20, 23, 24*). Some reports (*10*) of high mortality of birds have been attributed to poisoning by organochlorines. The bioconcentration of DDT and other organochlorine residues is apparently associated with the habitat and dietary habits of different species of birds (*3, 4, 9, 17, 23*). For example, earthworms are the principal source of DDT for robins (*1*). These pesticides enter the bodies of earthworms through soil, which is the largest reservoir of pesticide residues.

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Certain species of birds, because of their worldwide distribution, are considered good indicators of environmental pollution by pesticides (*16*). For example, crows have been used by many workers (*11, 15, 18, 21, 22*). The present report deals with DDT and BHC residues in wild pigeon (*Columba livia*), house crow (*Corvus splendens*), common pariah kite (*Milvus migrans*), Bengal vulture (*Gyps bengalensis*), and cattle egret (*Bubulcus ibis*). Farm-bred chickens were taken for comparison.

Materials and Methods

Wild pigeon, crow, kite, vulture, and cattle egret (three birds in each species) were obtained through commercial bird trappers during February and March 1980, from the urban area of Lucknow, a major city located 26°52′ north and 80°56′ east in the Indo-Gangetic plain. It is one of the most populated regions in the world, with a tropical climate. Chickens (average body weight 500 g) were purchased from the State Live Stock Farm, Lucknow. Within 24 hours of capture, birds were sacrificed, blood was collected in heparinized containers, and plasma was separated. Depot fat and internal body organs were excised. The average bird weights were 200, 305, 750, 4200, and 260 g for pigeon, crow, kite, vulture, and cattle egret, respectively.

ANALYSIS FOR ORGANOCHLORINE PESTICIDE RESIDUES

Blood Plasma—One milliliter blood plasma was mixed with 3 ml concentrated formic acid (98 percent pure) and extracted with *n*-hexane by shaking 1 hour. The *n*-hexane extract was washed with glass-distilled water and cleaned by concentrated H_2SO_4 treatment according to Dale et al. (*6*), as modified in the Arrhenius Laboratory, Analytical Chemistry, University of Stockholm, Sweden.

Body Tissues—Minced tissue (2 g) from body organs was thoroughly homogenized with 7 ml formic acid and transferred to a 50-ml conical flask. The homogenizing tube and pestle were washed twice with 5-ml portions of *n*-hexane, and the washings were collected in the flask. The homogenate was shaken in a 40°C water bath for 1 hour and then the solvent layer was withdrawn.

Minced depot fat (0.5 g) was homogenized with 3 ml formic acid and 5 ml *n*-hexane, transferred to a 50-ml conical flask, and treated as above.

The extracts of brain and body fat samples above were partitioned with acetonitrile (saturated with *n*-hexane) to remove fat. Pesticide residues were re-extracted in *n*-hexane. The solvent extract was passed through a column filled with anhydrous Na_2SO_4 and collected in a round-bottomed flask. The column was washed with 10 ml *n*-hexane and the washings were added to the original filtrate. The solvent extract was evaporated to dryness under reduced pressure and then re-dissolved in 5 ml *n-hexane*. Fractions (2 ml) were treated with 2 ml fuming H_2SO_4 and centrifuged, and the solvent layer was withdrawn.

Pesticide residues were determined by gas-liquid chromatography (Varian Aerograph Series 2400), using electron-capture detection (^3H), at the following operating conditions:

Carrier gas:	pure nitrogen passed through silica gel and molecular sieve to remove moisture and oxygen, respectively
Gas pressure:	65 psi
Gas flow:	40 ml/minute
Detector temperature:	200°C
Injector temperature:	190°C
Column temperature:	180°C
Column:	glass spiral column, 6 ft × ⅛ in. ID, coated with 1.5 percent OV-17 + 1.95 percent OV-210

Residue peaks were identified by thin-layer chromatography (TLC) on silica gel G-coated glass plates (*14*) and comparison with reference standards obtained from PolyScience Corp., Niles, Illinois. Further confirmation of the residues was done by chemical methodology (*12*) and column chromatography (*19*).

Recoveries of BHC isomers, DDT, and DDT metabolites (*p,p'*-DDE and *p,p'*-TDE) in the fortified samples of liver, brain, muscles, and body fat were between 70 and 94 percent. Sensitivity of the method was about 0.001 ppm for BHC isomers, aldrin, and *p,p'*-DDE and about 0.002 ppm for *p,p'*-DDT.

All reagents and chemicals used were high purity and were checked for interferences under the experimental conditions.

Results and Discussion

The concentrations of BHC and DDT residues in blood plasma, brain tissue, and depot fat are summarized in Tables 1 and 2. Levels of total BHC, γ-BHC (lindane), and total DDT in breast muscle, liver, heart, lung, kidney, and spleen of birds are shown in Figure 1. All values are expressed in terms of whole-tissue wet weight; results were not corrected for recovery.

DDT and BHC and their residues are widely distributed in the ecological system. Although BHC residues are excreted rapidly (*13*), slow accumulation does occur in the body tissues and body fat on chronic exposure. DDT and derivatives are quite stable and are resistant to enzymic action; thus, residues accumulate in biological tissues. The levels of DDT residues present in the body are occasionally taken as an index of contamination by DDT and DDT metabolites of the local environment. The general tendency appears to be that the smaller the

TABLE 1. *Range and geometric mean values of total BHC and γ-BHC (lindane) residues in blood plasma, brain, and depot fat of some wild birds and chickens*

	Residues, ppm whole-tissue wet weight					
	Blood Plasma		Brain Tissue		Depot Fat	
Bird	Total BHC	Lindane	Total BHC	Lindane	Total BHC	Lindane
Chicken	0.007 [1]	0.002	0.014	0.001	0.208	0.090
	0.006–0.010 [2]	0.002–0.003	0.008–0.025	0.001–0.002	0.121–0.310	0.075–0.120
	(0.008) [3]	(0.002)	(0.016)	(0.002)	(0.253)	(0.092)
Pigeon	0.048	0.016	0.316	0.020	—	—
	0.045–0.053	0.015–0.019	0.215–0.607	0.014–0.034	—	—
	(0.048)	(0.016)	(0.355)	(0.021)	—	—
Crow	0.030	0.011	0.246	0.014	21.815	6.550
	0.016–0.062	0.006–0.024	0.213–0.266	0.010–0.016	15.532–29.726	3.996–9.707
	(0.035)	(0.013)	(0.248)	(0.014)	(22.487)	(6.982)
Kite	0.060	0.028	0.093	0.012	5.468	2.876
	0.043–0.094	0.020–0.051	0.030–0.168	0.009–0.019	3.328–12.414	1.182–8.096
	(0.064)	(0.031)	(0.119)	(0.013)	(6.567)	(3.921)
Vulture	0.106	0.066	1.132	0.26	12.628	10.400
	0.066–0.143	0.044–0.080	0.759–2.047	0.142–0.683	6.510–19.823	4.951–18.964
	(0.112)	(0.068)	(1.247)	(0.335)	(13.980)	(11.966)
Cattle egret	0.004	0.048	0.126	0.053	6.309	4.873
	0.056–0.075	0.040–0.057	0.032–0.175	0.042–0.080	5.623–7.239	3.567–6.638
	(0.064)	(0.048)	(0.132)	(0.055)	(6.344)	(5.031)

[1] Geometric mean.
[2] Range.
[3] Arithmetic mean.

TABLE 2. *Range and geometric mean values of DDT and DDT metabolites in blood plasma, brain, and depot fat of some wild birds and chickens*

Compound	Residues, ppm Whole-Tissue Wet Weight					
	Blood Plasma	Brain	Depot Fat	Blood Plasma	Brain	Depot Fat
	CHICKEN			PIGEON		
p,p'-DDE	0.002	0.002	0.222	0.004	0.012	—
	0.002–0.003	0.001–0.002	0.190–0.250	0.003–0.006	0.005–0.021	—
	(0.002)	(0.002)	(0.226)	(0.004)	(0.014)	—
p,p'-TDE	0.002	0.001	0.055	0.001	0.001	—
	0.002–0.002	0.001–0.001	0.045–0.083	ND–0.002	ND–0.003	—
	(0.002)	(0.001)	(0.059)	(0.001)	(0.001)	—
p,p'-DDT	0.003	0.008	0.272	0.003	—	—
	ND–0.007	0.007–0.009	0.236–0.298	0.002–0.007	ND	—
	(0.004)	(0.008)	(0.274)	(0.004)	—	—
Total DDT	0.009	0.011	0.587	0.009	0.013	—
	0.005–0.012	0.009–0.012	0.550–0.670	0.005–0.014	0.005–0.023	—
	(0.009)	(0.010)	(0.590)	(0.010)	(0.017)	—
	CROW			KITE		
p,p'-DDE	0.035	0.035	18.595	0.100	0.038	23.108
	0.024–0.044	0.025–0.064	15.081–21.069	0.043–0.192	0.026–0.053	4.652–60.00
	(0.036)	(0.039)	(18.798)	(0.119)	(0.040)	(36.287)
p,p'-TDE	0.007	0.003	2.107	0.317	0.019	11.275
	0.006–0.010	ND–0.024	1.151–3.530	0.229–0.420	0.014–0.027	2.850–43.886
	(0.007)	(0.011)	(2.327)	(0.327)	(0.020)	(19.391)
o,p'-DDT	ND	ND	2.107	—	—	—
	—	—	ND–5.024	ND	ND	ND
	ND	ND	(2.289)	—	—	—
p,p'-DDT	0.018	0.004	12.563	0.044	0.003	4.496
	0.014–0.024	ND–0.030	4.488–63.421	0.027–0.074	ND–0.027	2.454–8.148
	(0.019)	(0.018)	(24.958)	(0.048)	(0.009)	(5.049)
Total DDT	0.066	0.063	44.486	0.522	0.076	44.986
	0.052–0.080	0.031–0.128	31.197–89.713	0.346–0.655	0.075–0.077	10.310–120.115
	(0.067)	(0.074)	(50.788)	(0.543)	(0.076)	(67.014)
	VULTURE			CATTLE EGRET		
p,p'-DDE	0.183	0.587	35.070	0.029	0.027	3.014
	0.106–0.245	0.298–1.052	20.297–53.948	0.026–0.033	0.018–0.035	2.131–3.049
	(0.196)	(0.655)	(37.878)	(0.030)	(0.028)	(3.110)
p,p'-TDE	0.209	0.386	39.543	0.004	0.004	6.991
	0.133–0.317	0.228–1.022	25.197–61.909	0.003–0.006	ND–0.014	2.289–23.478
	(0.222)	(0.499)	(40.736)	(0.005)	(0.006)	(10.710)
p,p'-DDT	0.039	0.095	6.162	0.003	—	3.979
	0.023–0.060	0.038–0.229	2.477–15.113	ND–0.006	ND	1.418–7.854
	(0.028)	(0.122)	(7.946)	(0.003)	—	(4.976)
Total DDT	0.479	1.204	87.945	0.041	0.038	17.048
	0.292–0.685	0.644–2.535	53.051–143.92	0.037–0.045	0.035–0.045	8.360–36.267
	(0.509)	(1.416)	(95.353)	(0.042)	(0.039)	(20.323)

animal the higher the ratio of food consumption per unit weight, regardless of food habits. Thus, within the same food habit group, smaller individuals are likely to ingest larger amounts of pesticide residues. Age and body size of the species are also important in influencing the accumulation of pesticide residues. However, food habits and the food-chain concentration mechanism are more important in determining the total body burden of pesticide residues.

Table 1 shows that levels of BHC and lindane present in blood plasma, brain tissue, and depot fat of kite and cattle egret are comparable. Relatively high levels of total BHC were detected in crow and pigeon brain, as compared with blood plasma levels. Vulture (a carrion-feeder) contained 1.25 ppm total BHC and 0.34 ppm lindane in the brain, which are many times higher than the concentrations present in other species of birds studied. As high as 29.73 ppm total BHC was recorded (average, 22.49 ppm) in crow depot fat.

BHC levels in liver, heart, lung, and kidney were generally high in pigeons and crows (Figure 1). Breast muscles and spleen of vulture exhibited relatively high accumulation of BHC. Kite and cattle egret showed generally low concentrations of these compounds in tissue. Compared with other isomers of BHC, lindane accumulation was high in the tissues of vulture and cattle egret. However, the α-isomer accounted for the major burden of this compound in brain tissue of all birds studied. A single specimen of pigeon ovary examined in the present study contained 1.21 ppm total BHC and 0.31 ppm lindane. Residue levels found in the tissues and blood plasma of chicken were relatively low.

Until 1962 (*8*), carcasses of random samples of birds had been analyzed for aldrin, dieldrin, or DDT. DDE is the primary breakdown product of DDT and is universally distributed; exposure to this compound is essentially a continuous one. Residues of parent compound, DDT, and the metabolite TDE are less frequent. Results of

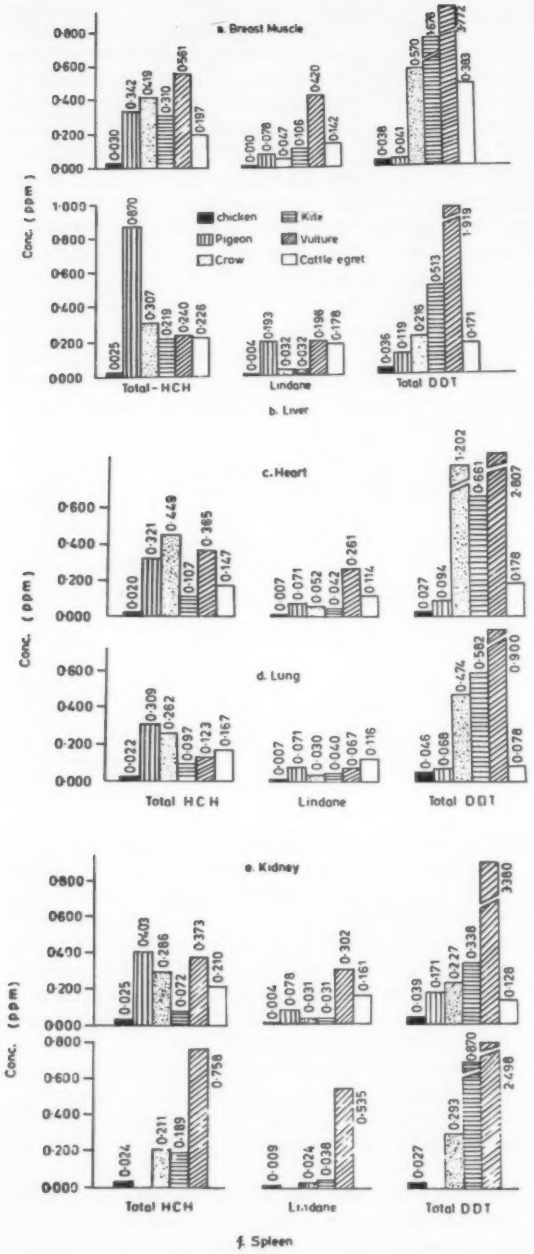

FIGURE 1. *Total BHC, lindane, and total DDT in internal body organs of different species of birds.*

the present study show a positive correlation of DDT concentration in blood and brain, as well as in blood and depot fat, as suggested by several workers (*2*, *5*). Levels of DDT and DDT metabolites in blood plasma, brain, and depot fat are presented in Table 2, and total DDT levels in the rest of the tissues are given in Figure 1.

Few samples of lungs, spleen, and depot fat of crow showed measurable levels of *o,p'*-DDT. *p,p'*-DDT was not detected in lung, kidney, and brain tissue of pigeon and cattle egret. *p,p'*-TDE was not found in the lungs of these birds. Relatively high levels of *p,p'*-DDE were observed in different body organs of cattle egret, compared with those present in other birds examined. More than 70 percent of total DDT was present as *p,p'*-DDE in most body organs of pigeon. An equivalent concentration of *p,p'*-DDT and comparatively low levels of *p,p'*-TDE in body organs of crow, and to some extent chicken, indicate that the exposure is rather continuous from different environmental sources. Vulture and kite contained high concentrations of *p,p'*-DDE and *p,p'*-TDE, compared with the levels of parent compound (*p,p'*-DDT).

Total DDT levels were 0.59, 20.32, 50.79, 67.10, and 95.35 ppm in depot fat of chicken, cattle egret, crow, kite, and vulture, respectively. In other tissues, total residues of DDT generally occurred in the following order: chicken < pigeon < cattle egret < crow < kite < vulture. Total DDT detected was 0.01, 0.01, 0.04, 0.07, 0.054, and 0.51 ppm in blood plasma and 0.01, 0.02, 0.04, 0.07, 0.08, and 1.42 ppm in brain tissue of chicken, pigeon, cattle egret, crow, kite, and vulture, respectively. The sample of pigeon ovary analyzed contained 1.01 ppm total DDT.

DDT and its metabolites show a consistent biomagnification in wild birds, presumably through the food-chain concentration mechanism. Flesh-eating birds had shown higher body burdens of DDT than non-flesh-eating ones. Thus, birds of the upper trophic zone in the food chain show higher bioaccumulation of DDT residues. DDT levels present in birds are perhaps a reflection of the environmental status of the habitat and food choices of particular avian species.

Acknowledgment

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Cadmium, Lead, Mercury, Arsenic, and Selenium Concentrations in Freshwater Fish, 1976–77—National Pesticide Monitoring Program

Thomas W. May [1] and Gerald L. McKinney [2]

ABSTRACT

As part of the National Pesticide Monitoring Program, the Fish and Wildlife Service, U.S. Department of the Interior, collected freshwater fish during 1976–77 from 98 monitoring stations and analyzed them for residues of cadmium, lead, mercury, arsenic, and selenium. Range and geometric mean values in mg/kg wet weight follow: Cd, 0.01–1.04, 0.07; Pb, 0.10–4.92, 0.32; Hg, 0.01–0.84, 0.11; As, 0.05–2.92, 0.27; Se, 0.05–2.87, 0.56. An arbitrary 85th percentile was calculated for concentrations of each element in fish to identify monitoring stations having fish with higher-than-normal concentrations: Cd, 0.11 mg/kg; Pb, 0.44; Hg, 0.19; As, 0.38; Se, 0.82. Log-transformed mean concentrations in fish from 1976–77 monitoring stations are compared with means from the same stations in 1972 (Cd, Hg, Pb, As, Se) and 1973 (Se) to depict temporal trends in whole-body concentrations: Cd, significant decline; Pb, no significant difference; Hg, significant decline; As, significant increase; Se, no significant difference. Because of changes in laboratories and analytical procedures, these conclusions should be used cautiously as trend information. Production, consumption, and disposal of cadmium, lead, mercury, arsenic, and selenium are discussed as potential environmental sources of the elements to the aquatic environment. Specific environmental sources are suggested for monitoring stations having trace element levels exceeding calculated 85th percentiles.

Introduction

The National Pesticide Monitoring Program (NPMP) is a Federal program established to monitor nationwide environmental contaminants in air, soil, water, humans, plants, and animals. United States government agencies participating in NPMP are the U.S. Environmental Protection Agency (EPA); Geological Survey, U.S. Department of the Interior; Food and Drug Administration, U.S. Department of Health and Human Services; U.S. Department of Agriculture; and Fish and Wildlife Service, U.S. Department of the Interior.

The Fish and Wildlife Service (FWS) is responsible for monitoring selected environmental contaminants in freshwater fish. Although primary emphasis has been placed on organic contaminants, selected trace elements have been determined intermittently. In 1969, 3 composite samples, each of a different species and consisting of 3 to 5 whole adult fish, were collected from 50 sampling stations and analyzed for mercury (*20*). Cadmium, lead, and arsenic were added to the program in 1971, and selenium was added in 1972. Sample collections included a replicate for each species in 1971, but for only one of three species from each station in 1972. In 1973, all samples were analyzed for selenium, but only selected samples were analyzed for mercury, arsenic, lead, and cadmium. The 1971–73 analyses were conducted by the Denver Wildlife Research Center, FWS (*87*). Samples were collected from 97 stations in 1974, but no trace elements were analyzed. Sample collections were suspended during the 1975 sampling year to enable a technical and administrative review of fish-monitoring activities. The freshwater fish-monitoring program was reviewed internally and restructured, and responsibility for NPMP in FWS was shifted to the Columbia National Fisheries Research Laboratory (CNFRL).

In 1976, collection stations were increased from 100 to 117 to include a more extensive coverage of the Great Lakes. Collections were then modified to include duplicate composite samples of a bottom-dwelling species and one composite of a representative predatory species at each station. A list of acceptable bottom-dwellers and predator species, listed in order of priority in Table 1, was developed. The collections included 146 samples from 52 stations in 1976 and 163 samples from 54 stations in 1977 (Figure 1). The purpose of this report is to present and interpret heavy metals data gathered for the NPMP during 1976–77.

Environmental Sources of Heavy Metals

Production, consumption, and disposal processes often result in the transport of trace elements to the aquatic environment. The U.S. EPA has established priorities for trace element contamination problems and threats to resources by including 13 trace elements on the

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TABLE 1. *Sequential priority for selection of bottom-feeding and predator species of fish, as established by the National Pesticide Monitoring Program* [1]

BOTTOM FEEDERS (commercially or recreationally significant, if feasible)

(1) Carp (*Cyprinus carpio*)
(2) Common sucker (*Catostomus commersoni*) or other members of the sucker family
(3) Channel catfish (*Ictalurus punctatus*) or other members of the catfish family
(4) Other, with justification

PREDATORS (should be an important sport fish)

(1) Cold water stations: rainbow trout (*Salmo gairdneri*), brown trout (*Salmo trutta*), brook trout (*Salvelinus fontinalis*), lake trout (*Salvelinus namaycush*)
(2) Warm-water stations: largemouth bass (*Micropterus salmoides*), or other member of the sunfish family, such as crappie (*Poxomis* sp.), bluegill (*Lepomis macrochirus*), etc.
(3) Cool-water stations: walleye (*Stizostedion vitreum*) or other members of the perch family
(4) Other, with justification, but must be representative of the drainage system

[1] From National Pesticide Monitoring Program, Freshwater Fish Collection Instructions, internal memorandum issued annually to FWS Regional Pesticide Specialists.

priority pollutants chemicals list. In this section, authors attempt to link production-consumption practices associated with cadmium, lead, mercury, arsenic, and selenium to environmental sources to clarify the rationale for monitoring concentrations of these trace elements in freshwater fish.

CADMIUM

Cadmium has a close geochemical association with zinc, and natural geochemical sources of cadmium are linked with zinc deposits occurring as massive-sulfide and sulfides in strataform carbonates. Nearly all domestic cadmium is produced as a by-product of zinc concentrates and imported zinc smelter flue dusts (*66*). The primary domestic producers of cadmium in 1978 were AMAX Zinc Co., Inc., Sauget, Illinois; ASARCO Inc., Corpus Christi, Texas, and Denver, Colorado; Bunker Hill Co., Kellogg, Idaho; National Zinc Co., Bartlesville, Oklahoma; New Jersey Zinc Co., Palmerton, Pennsylvania; and St. Joe Zinc Co., Monaco, Pennsylvania.

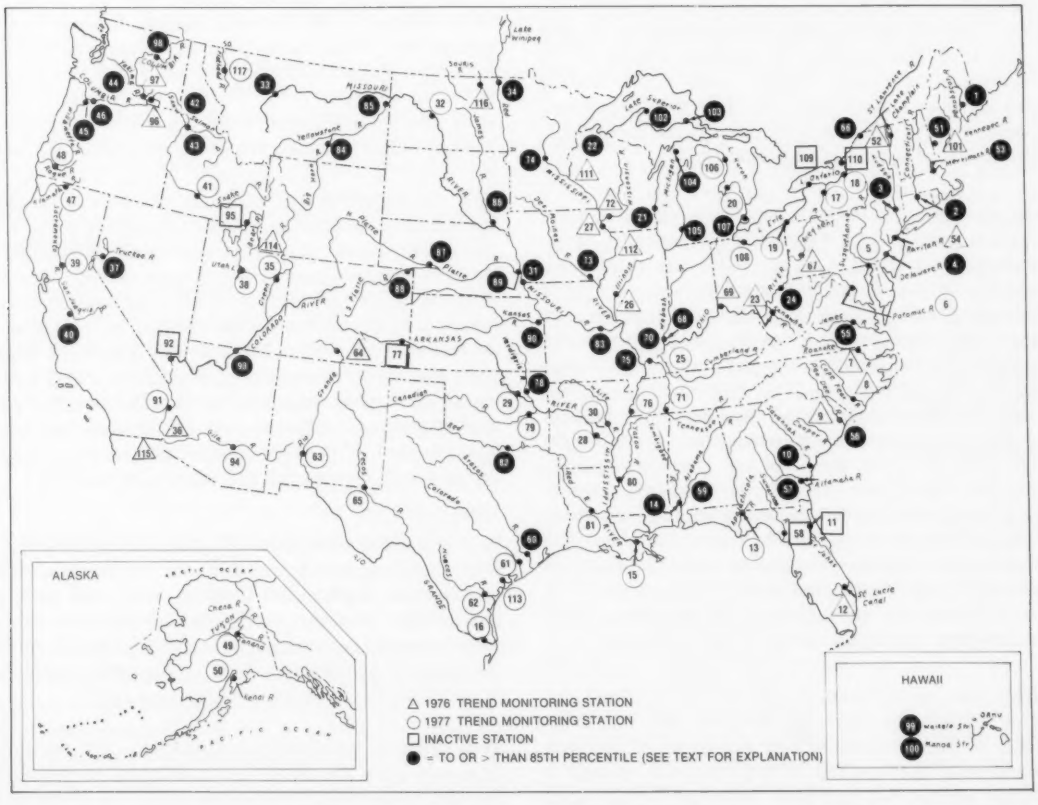
ALASKA
HAWAII
△ 1976 TREND MONITORING STATION
○ 1977 TREND MONITORING STATION
□ INACTIVE STATION
● = TO OR > THAN 85TH PERCENTILE (SEE TEXT FOR EXPLANATION)

FIGURE 1. *Stations sampled for determination of some metals in freshwater fish as part of National Pesticide Monitoring Program, 1976–77.*

Of the domestic consumption of cadmium, 95 percent is divided into five principal uses: plating, pigments, alloys, batteries, and plastic stabilizers. Electrically or mechanically plated hardware used in vehicles and other equipment accounted for 40–45 percent of domestic cadmium consumption in 1978. The production of red, orange, yellow, and maroon pigments consumed 15 percent of the supply, and most of the rest was used in nickel-cadmium batteries, special-purpose alloys, and compounds providing heat and light stability to plastics, particularly polyvinyl chloride (*63*).

In addition to smelter production of cadium metal, primary producers have emphasized production of various cadmium compounds in recent years (*63*). Cadmium is used in the manufacture of pesticides for control of moles and plant diseases affecting residential lawns and golf courses (*63*).

Cadmium is released to the environment primarily from four sources: the electroplating industry; the smelting and refining of zinc, lead, and copper; the application of phosphate fertilizer; and surface mine drainage (*15, 80*). The tendency of cadmium to concentrate in sediments (*15*) may result in a persistent source of the contaminant to various trophic levels in the aquatic environment. Studies indicate that fish accumulate cadmium from the water and through the food chain; both modes of uptake can be toxic (*80*).

The electroplating industry uses cadmium plating for corrosion protection. Wastewater is generated from countercurrent rinses, rinses following chromating, dumping of the chromating solution, and purging of the plating-recovery loop (*82*). Plant outlet pipes often lead to municipal sewage treatment plants, where about 50 percent of the cadmium remains with sewage sludge, and the rest is discharged (*81*). Incineration of sewage sludge volatilizes cadmium, whereas deposition in landfill areas subjects streams and groundwater to contamination. Snowmelt from the roofs and grounds of plating firms can contain more than 1 ppm cadmium, originating as particulate droplets of plating solutions exhausted from the interiors of the firms by fans and accumulating on the roofs or walls or the ground below the discharge point (*55*). Perlmutter and Lieber (*70*), who traced the spread of cadmium from a plating plant, found groundwater containing up to 10 ppm cadmium.

When zinc ores are roasted, cadmium is volatilized and partly collected as fumes or flue dust. The rest is released to the atmosphere and deposited in the area surrounding the smelter (*15*). The soil around a smelter facility that had been operating for 80 years was contaminated with cadmium within a radius of at least 10 miles (*76*). Flue dust returned to the smelter is often stored in waste cinder banks, which are a source of pollution due to leaching and erosion from rainwater (*78*). Suspended sediments containing up to 0.1 percent cadmium have been reported in streams from high runoff areas near copper and zinc smelters (*78*). Inland smelters and mills generally have extensive slag heaps and tailing ponds along streams for waste disposal. Estuarine and river refineries generally dispose of wastes directly into the water through outfalls.

Phosphate ores used in fertilizer manufacture may contain from 9 to 130 ppm cadmium (*77*). Runoff from agricultural areas where phosphate fertilizers are used could result in substantial cadmium loading to the aquatic environment.

The primary sources of cadmium contamination during mining are the emission of particulates and the leaching of cadmium from the overburden. Ores are enriched by flotation techniques to yield a 40–60 percent cadmium metal product. Because the cadmium concentration in a mine is only a few percent, most of the original ore mass becomes tailing waste. As much as 18–36 percent of the cadmium may be retained in the tailings (*15*). Mine waters from sulfide ores can contain more than 40 ppm cadmium, but levels of 0.1 to 2 ppm are more common (*15*). Draining from coal mining areas also poses a threat: Eight bituminous coals from Kentucky and Pennsylvania had 1-2 ppm cadmium (*30*).

Low concentrations (< 0.1 ppm) of cadmium in fish are deceptive, because cadmium is extremely toxic and cumulative. Benoit et al. (*6*), who measured cadmium levels in various tissues of brook trout exposed to cadmium in water for up to 38 weeks, reported that the kidney accumulated the highest concentration, followed by the liver and gills. Exposed fish placed in fresh water lost cadmium rapidly from gill tissue but did not lose it from either the kidney or liver. In a similar 40-week exposure of rainbow trout to cadmium, made by Kumada et al. (*29*), almost no cadmium was lost from the kidney of exposed fish returned to fresh water.

LEAD

Lead is a major constituent of certain geological formations, including stratabound deposits, volcanic-sedimentary deposits, replacement deposits, veins, and contact metamorphic deposits. Lead ore deposits commonly contain the sulfide mineral galena (PbS), which is often associated with sphalerite (ZnS), pyrite (FeS_2), chalcopyrite ($CuFeS_2$), and other sulfur salts (*64*).

Most domestic primary lead production (88 percent) originates from the limestone or dolomite stratabound deposits of southeastern Missouri (*66*). The silver-lead vein system of Idaho's Coeur d'Alene District provides 8 percent of domestic primary lead, and the rest is

provided by replacement deposits in Colorado (3 percent) and Utah (1 percent). In 1978, primary lead was smelted and refined at seven U.S. plants (*65*): four ASARCO plants in El Paso, Texas; East Helene, Montana; Omaha, Nebraska; and Glover, Missouri; and AMAX in Buick, Missouri; St. Joe Minerals in Herculaneum, Missouri; and Bunker Hill in Kellogg, Idaho. Because of the relative ease of reclaiming the metal, old scrap lead (secondary lead) accounted for 51 percent of domestic consumption in 1978. More lead is now produced from secondary sources than from domestic ores (*66*).

The transportation industry is the major end user of lead: 51 percent is consumed in storage batteries and 15 percent in lead alkyl compounds that are used as gasoline antiknock additives. The electrical industry (8 percent consumption) has long depended on lead for cable coverings where corrosion or moisture problems may exist. Because of its toxicity, lead is no longer used in interior paints and has been largely replaced in exterior paints by zinc and titanium pigments. Lead pigments are still the preferred base material for corrosion protection in structural and highway components (6 percent of total consumption). In the ammunition industry, lead remains the major metal in shot and small-caliber bullets (4 percent consumption). The construction industry (3 percent consumption) is using increasing amounts of lead as a sound barrier in partitions and ceilings, as well as in roofing, piping, flashing, and caulking. Various other industries use lead for many different purposes and together account for 13 percent of consumption (*66*).

Lead enters the environment from several sources. The major source of lead emissions (88 percent) is the combustion of leaded gasoline. Although environmental restrictions, initiated in 1972 to control air pollution, have reversed the growth in use of lead antiknock additives, unleaded gasoline accounted for only 33 percent of the gasoline sold in 1978 (*83*). The average lead content in pooled (leaded and unleaded) gasoline in 1972 was 1.2 g/gal. Under cruise conditions, lead is emitted from automobile exhausts in the form of small particles, most of which are < 1 μm in mass median equivalent diameter (*32*). Such a small particle size increases the residence time of lead emitted into the atmosphere and, consequently, dispersion from the point of emission. The small size of the particles emitted is generally characteristic of urban lead aerosols, and concentration of lead in ambient air is strongly correlated with automobile traffic density (*39*). Thus, atmospheric fallout and surface runoff of lead into streams and rivers should be most intense where waterways flow through metropolitan areas.

Smelting and mining of lead, zinc, and copper have caused marked environmental contamination problems, even though lead emissions from these sources are small relative to vehicle exhausts. Sediments containing up to 17 percent lead by weight have been found below zinc and copper extractive industries, in streams used for irrigation and drinking water (*78*). The pollution hazard is greatest where there is erosion of waste cinder banks, tailings, and slag heaps. Although most large smelters are equipped with efficient dust and fume collection systems that claim 98 percent recovery (*64*), the recovered flue dusts are sometimes stored in unprotected waste cinder banks, where leaching and erosion by rainwater result (*78*). Despite efficient stack collection systems, aerial fallout of lead has resulted in severe local contamination. Leaves of post oak (*Quercus stellata*) and shortleaf pine (*Pinus echinata*) within 0.5 mile of a lead smelter in Missouri contained levels of lead as high as 8,125 and 11,750 ppm (*7*). Samples of various plant species containing normal lead concentrations could be obtained only beyond a 20-mile radius from the smelter-mining-milling complex. The deaths of 20 horses prompted the analyses of samples of forage grass in the vicinity of another Missouri smelter, in which concentrations as high as 14,700 ppm lead were reported (*7*).

Lead mines associated with limestone or dolomite stratabound deposits (Missouri's Old and New Lead Belts) must pump out 5,000–7,000 gal/min of groundwater in order to operate. The relatively clear water is typically cycled through the mill and flotation concentrators and ends up containing mud, organic flotation agents, and other wastes. This effluent is discharged into valleys formed by dams of coarser mill tailings, and the final effluent is the tailing pond outfall. In the New and Old Lead Belt mining-milling areas, tailing pond outfalls have resulted in the deposition of a dark lead-bearing dolomite mud on stream bottoms; the mud in turn is covered by a gray algal-bacterial slime. Benthic fauna were found to be intolerant of the dolomite mud covering (*78*).

Other environmental sources of lead are landfills or dumps, fly ash from coal-burning power stations, coal combustion, sewage sludge, and application of pesticides containing lead (*47*). Small emissions occur from lead oxide manufacturing and fuel oil combustion (*39*). Coal mining could contribute significant quantities of lead to the environment during flood erosion (*78*).

Upon entering natural waters, most lead is precipitated to the sediment bed as carbonates or hydroxides (*80*). Laboratory studies have shown that lead compounds can be transformed to tetraalkyllead, but the exact mechanism is still unclear. Wood et al. (*90*) proposed a Type I microbial methylation reaction for lead, where the high

redox potential of the Pb IV/Pb II redox couple causes Pb IV to act as an attacking electrophile. Subsequent heterolytic cleavage of the Co–C methylcobalamin bond results in the transfer of a carbanion methyl group to the more oxidized form of the element. Jarvie et al. (*25*), however, were unable to achieve methylation of trimethyllead salts and lead nitrate by this microbial pathway and, instead, proposed a chemical mechanism for conversion of the compounds to tetramethyllead in active anaerobic sediments. Other workers (*51*) have demonstrated the methylation of lead(II) compounds to tetramethyllead by microorganisms, which suggests other routes of methylation besides the mentioned microbial and chemical routes. It is not now known whether lead, like mercury, can accumulate through the food chain as an alkylated entity. Because divalent lead is the principal form accumulated by aquatic animals, the possibility of methylation of ionic lead in vivo cannot be disregarded (*80, 53*). Tetraalkyllead compounds have been found in various marine tissues (*53*).

MERCURY

Mercury has an impressive list of uses encompassing many different types of industry and has almost 3,000 distinct applications (*12*). The largest end user in the United States is the electrical apparatus industry, which accounts for 42 percent of total consumption and includes the manufacture of mercury batteries and alkaline energy cells, vapor discharge lamps, rectifiers, and switches. The second greatest use (16 percent) is in the electrolytic preparation of caustic soda and chlorine (chloralkali industry), where the continuous-flow mercury cathode cell still accounts for about 20 percent of total chloralkali-producing capacity (*4*). Mercury consumption in the United States for chloralkali purposes has been reduced sharply since the 1960's for at least three reasons: a decrease in the number of new mercury cell chloralkali plants, modification of existing plants to reduce mercury losses, and conversion of some plants to the diaphragm process. The paint industry consumes 13 percent of the mercury used in the United States, mostly for mildew proofing (*58*). Industries manufacturing industrial control instruments consumed about 8 percent of U.S. mercury supplies in the manufacture of switches, relays, gauges, pump seals, and valves. Other uses, which account for about 15 percent of total mercury consumption, include those in agriculture, dentistry, general laboratory applications, and pharmaceuticals (*4*). Mercury consumption for pesticide use in agriculture is down sharply from the late 1960's, but a relatively small number of mercury pesticide formulations are currently available (*5, 58*). The U.S. paper and pulp industry no longer uses mercury as a slimacide, but still may be consuming mercury at the combined chloralkali-pulping operations (*12*).

There are two primary ways mercury reaches the aquatic environment: pre-1975 chloralkali operations and pre-1972 paper-pulping operations. Although the introduction of mercury to the environment from these industries is now relatively small, stream and lake sediments contaminated from discharges 10–15 years ago are a persistent mercury source, and methylation by anaerobic microbes initiates bioconcentration and food chain bioaccumulation (*12*). Seepage from some waste disposal areas of closed chloralkali plants continues to contaminate streams and reservoirs (*56*). Higher-than-background mercury sediment concentrations have been found more than 100 miles downstream from a synthetic fiber operation that stopped using mercury 27 years ago (*3*).

Although U.S. mercury consumption and industrial mercury loss have been reduced from early 1970 levels, mercury contamination associated with increased coal and crude oil production may pose future problems. Fossil fuels contain from 10 ppb to several ppm mercury, depending on the coal type (*12*). Inasmuch as the United States is preparing for a dramatic intensification of coal mining, combustion, and conversion, it appears likely that an environmental mercury problem will be present for some time to come.

Bacteria present in most natural waterways can convert mercury to methylmercury. Ridley et al. proposed a Type I microbial methylation reaction for mercury that is very similar to that already mentioned for lead (*49, 90*). Most of the mercury in fish exists as methylmercury derived largely from food. Some authors have suggested that water, as well as food, is a major source of methylmercury in fish (*80*).

ARSENIC

Arsenic occurs in association with complex base-metal ores, chiefly those of copper, lead, gold, and to a lesser extent, cobalt and tin (*60, 66*). The element is a minor constituent of those ores and is regarded as a troublesome impurity in smelting and refining of base metals. The recovery of arsenic in residues from fumes, skimmings, and flue dusts involves sophisticated technology and is costly and relatively inefficient. As a result, the refinery incentive for arsenic recovery is closely correlated with concurrent economics and market conditions. In 1978, all domestic production of arsenic was confined to the copper smelting-refining complex of the American Smelting and Refining Company (ASARCO) in Tacoma, Washington. Anaconda, another large Northwestern copper smelting company located in Butte, Montana, has arsenic-refining facilities that have remained unused for the past several years. Because domestic arsenic production is so limited and availability is so closely tied to prevailing copper prices, the United

States historically has met most of its requirements for arsenical compounds by importation (*60*). For example, domestic production supplied only 10.5 percent of total U.S. arsenic demand in 1973 and about 50 percent in 1978 (*60, 66*).

Almost all arsenic (97 percent) enters end-product manufacturing in the form of white arsenic or As_2O_3. The other 3 percent is in the metallic form and is used as an additive in specialized lead and copper alloys. Eighty-two percent of white arsenic is consumed in the manufacture of agricultural pesticides, such as lead arsenate, calcium arsenate, sodium arsenite, and organic arsenicals that are used as insecticides, herbicides, fungicides, algicides, desiccants, and defoliants (*60*). Organoarsenical compounds include cacodylic acid, disodium methanearsonate (DSMA), monosodium methanearsonate (MSMA), and sodium cacodylate (*5, 60*). The only extensive use of arsenic that is not based on its toxicity is in the glass industry, where it is used as a decolorizer and as a constituent of opalescent glass and enamels. Other small uses (*60*) are in the paint industry (pigments), pyrotechnics (constituent of fireworks), pharmaceuticals (treatment for skin disorders and sleeping sickness), electronics (diodes, transistors, and lasers), and the metals industry (as an additive to ferrous alloys to increase cast iron strength).

Arsenic enters the aquatic environment by four primary routes: (1) Dissemination by air pollution. Because of the complexity, inefficiency, and expense of removing arsenic from smelter stack gases, the element has become a major air pollution problem in states having smelting-refining operations (*60*). Coal combustion is another important source of arsenic to the air. (2) Smelter solid waste disposal. Because no domestic metallurgical plants, except ASARCO, process commercial arsenic, the disposal of fumes, skimmings, and flue dusts could constitute a solid waste pollution problem affecting both soil and water. (3) Arsenical pesticides. Continued use is expected for many years to meet the demand for effective pest control in the face of expanding agricultural production (*60, 66*). (4) Geologic. Because arsenic is found in association with specific geologic formations of volcanic origin, ground and surface waters in some areas of the western United States have high arsenic levels (*31*).

Arsenic occurs in natural waters primarily in the arsenate-arsenite forms (*27, 79*). Inorganic forms of arsenic can be methylated by various microorganisms, including fungi, methanogenic bacteria, yeasts, and unicellular algae (*43*). Evidence suggests that arsenic salts in the lower oxidation states perform a free-radical attack (homolytic cleavage) on the Co–C bond of methylcobalamin or nucleophilic attack on *S*-adenosylmethionine, as well as on methylcobalamin (*49, 90*). Fish apparently can biosynthesize organoarsenic compounds within the gastrointestinal tract (*34, 43*). However, the main source of arsenic for fish is primarily organoarsenic compounds that are synthesized at lower stages in the food chain (*34*). Generally, arsenic is not biomagnified in aquatic food chains. Penrose et al. (*44*) suggested that organisms at each trophic level convert inorganic arsenic to a detoxified organic form, organisms at the next higher trophic level then rapidly excrete the ingested organic arsenic, precluding food chain bioaccumulation.

SELENIUM

In 1978, all primary selenium was produced as a by-product from the processing of copper refinery slimes to recover gold, silver, and tellurium. Three copper refineries (AMAX Copper, Inc., Carteret, New Jersey; ASARCO Inc., Amarillo, Texas; and Kennecott Copper Corp., Magna, Utah) accounted for all domestic production of selenium (*62, 66*). Secondary production, or recycling, was limited; only about 1 percent of the 1978 consumption was recovered from xerographic and rectifier scrap and chemical waste products. Domestic consumption of selenium decreased steadily from 1974 to 1977 and increased slightly in 1978 (*66*). Major end uses of selenium in 1978 (*66*) were in electronic and photocopier components (35 percent), glass manufacturing (30 percent), and chemicals and pigments (25 percent). The electronics industry used selenium in dry-plate rectifiers for many years, but silicon, germanium, and cadmium have largely replaced it in these applications. The use of metal drums coated with photoconducting amorphous selenium in the dry photographic process of xerography has become a major end use of the metal. Selenium has the property of converting light energy directly into electrical energy—a property that has enabled the development of numerous photocell devices, such as photographic exposure meters and solar batteries (*41*).

The glass and ceramics industry adds selenium to glass melt to control final product color. Selenium is used to neutralize green tinting caused by iron impurities, resulting in the manufacture of clear glass. Addition of more selenium to the melt produces a pink-to-ruby red glass. Its use in dark-colored glass in buildings and vehicles to reduce glare and heat transfer is increasing.

A large number of selenium compounds have commercial uses, ranging from semiconductor research to anti-dandruff agents in shampoos. Much of the selenium consumed by the chemical industry is used to prepare pigments containing selenium. A major class of pigments is the cadmium sulfoselenide compounds, which have superb resistance to sunlight, heat, and chemical attack (*41*).

The primary sources of selenium in the environment are geologic and industrial. Selenium closely resembles sulfur chemically, and sulfur or sulfide deposits of bismuth, copper, iron, lead, mercury, silver, and zinc sometimes contain as much as 20 percent selenium (*68*). Other sulfate minerals, such as barite and jarosite, contain selenium, and native sulfur can contain more than 0.1 percent selenium. Other geologic formations containing selenium include sandstones, limestones, and shales. Sandstones containing > 100 ppm selenium have been found in Wyoming (*17, 91*). The Niobrara formation, a limestone region of South Dakota, contains > 40 ppm selenium in chalky shales and marls. Phosphate rocks associated with limestone may contain from 1 to 300 ppm selenium, suggesting the occurrence of selenium in phosphate fertilizers. Of the sedimentary rocks, shales have been mainly responsible for cases of selenium poisoning in animals in the United States. For example, vegetation in some areas of the Pierre Formation near the Missouri River in southern South Dakota has potentially toxic selenium concentrations. These shales are considered highly seleniferous and have selenium levels ranging from 1 to > 30 ppm (*41*).

Industry releases selenium to the environment through combustion of coal and fuel oil, nonferrous smelting and refining processes, metal refining, and glass manufacturing. Domestic coal averages 3.2 ppm selenium (*46*). Average selenium concentrations are 1.3 ppm in lignite coal and 2.08 ppm in central and western U.S. coals (*42*). In one study, about 53 percent of the selenium in coal was emitted to the atmosphere during combustion, either as volatilized selenium or in association with fly ash particles too small to be trapped by precipitators (*41*). Coal combustion accounted for 62 percent of the total industrial emission of selenium in 1970 (*41*). EPA found that crude oil contained an average of 0.4 ppm selenium (*41*), and Hashimoto et al. (*19*) reported averages of 0.92 ppm in raw petroleum and about 1.0 ppm in heavy petroleum. Smelting and refining of nonferrous metals produces slag heaps and tailing dumps containing high concentrations of selenium. Thus, solid wastes from metal mining and milling may be a more serious source of selenium pollution than is atmospheric fallout from base metal smelting and refining (*41*). Selenium emissions are high in glass manufacturing, because the high temperature of the glass melt volatilizes selenium (*41*).

Attempts to correlate atmospheric concentrations of selenium with the location of industrial selenium emissions have generally met with only limited success. For example, Traversy et al. (*24*) found the highest selenium concentrations in precipitation samples at or near highly industrialized locations in the Great Lakes region, and Copeland (*10*) showed that selenium concentrations in Lake Michigan zooplankton increased near Chicago. In these two situations, the effects of selenium from industrial emissions appeared to be localized, and natural sources of selenium may generally be more important than anthropogenic ones (*41*).

Ingestion may be the most important mode of selenium uptake by aquatic biota, but more research is needed to confirm this possibility (*56*). Phillips and Russo (*80*) concluded that the poor survival of stocked fish in a highly seleniferous Colorado lake was due to the accumulation of excess selenium through the food chain. Several species of molds and microorganisms can methylate selenium (*41*). Little is known, however, about selenium methylation pathways in the aquatic environment.

Methods and Materials

SAMPLE COLLECTION

Fish were collected by FWS biologists, state fish and game personnel, and local commercial fishermen, who used a variety of nonchemical collecting techniques (e.g., trapping, electrofishing, seining). After the total length and weight of each fish had been determined, the sample composites were separately wrapped in aluminum foil, frozen, and shipped to CNFRL.

Fish from frozen composites were reduced to ice-cube-sized blocks with a Hobart Model 5212 food service band saw. Blocks were passed twice through a large (Hobart 1 hp Model 4822) or small (Hobart ¼ hp Model 4612) meat grinder, depending on total composite size and weight. Between sample homogenizations, the band saw and disassembled grinder components were washed with hot water or steam and rinsed with deionized water. About 400-g portions of ground fish were placed in an acid-cleaned glass jar with a Teflon-lined cap and stored in a freezer. In 1976, frozen 10-g portions of 83 samples representing 44 stations were sent to the EPA Region VII Laboratory in Kansas City for digestion and analysis. All samples collected in 1977 were prepared and analyzed at CNFRL.

DIGESTION AND ANALYSIS OF SAMPLES COLLECTED IN 1976

Arsenic, Cadmium, and Lead—Five grams of thawed fish homogenate were placed in 10-in. Technicon digestor tubes. Ten milliliters of concentrated HNO_3 was added to each tube, and the sample–acid mixture was held at room temperature for 1 hr to reduce foaming when heat was applied. Samples were heated on a Technicon Model BD-40 block digestor at 150°C for 60 minutes and at 250°C for 90 minutes. The gradual increase from room temperature to 150°C allowed the samples to dissolve with little foaming; heating to 250°C was required to overcome reflux action at the constriction of the digestor tube. Samples were heated to dryness

to decompose lipid material. If the sample was black when dryness was reached, it was removed from the digestor, cooled, treated with an additional 10 ml concentrated HNO_3, and returned to a cold digestion block set for 90 minutes at 250°C. Subsequent addition of 10-ml portions of acid was continued until the appearance of a white residue indicated complete digestion. The white residue was dissolved with 10 ml 10 percent HNO_3 at 90°C. Dilution to 50 ml with deionized water provided a final acid matrix of 2 percent HNO_3. This procedure allowed simultaneous preparation of up to 40 tissue samples, with only HNO_3 as the oxidizing acid. Unfortunately, the recovery of selenium by this method was incomplete, precluding the use of selenium data originating from the 1976 samples.

Fish tissue digestates (2 percent HNO_3) were analyzed by a Jarrell-Ash Model 975 Atomcomp inductively coupled argon plasma (ICAP) optical emission spectrophotometer. Samples were introduced in a cross-flow nebulizer at 1.4 ml/min by a Gilson eight-channel pump. An autosampler maintained sample flow at 30 samples/hr. Other pertinent ICAP parameters follow:

Incident RF power:	1.1 kW
Reflected RF power:	20 W
Observation height:	15 mm above load coil
Sample argon flow rate:	0.5 l/min
Coolant argon flow rate:	18 l/min

Detection limits for arsenic, cadmium, and lead were 0.25, 0.05, and 0.10 mg/kg, respectively.

Mercury—One gram of sample was placed on the bottom of a dry BOD bottle; 1 ml each of concentrated H_2SO_4 and HNO_3 was added, and the bottle was placed in a water bath at 58°C until the tissue was completely dissolved (30–60 minutes). The bottle was cooled to 4°C in an ice bath, and 1 g $KMnO_4$ crystals was added to maintain oxidizing conditions. The digestate was stirred, loosely capped, and held overnight at room temperature (*75*).

The cold-vapor atomic absorption method for mercury analysis (*75*) can be summarized as follows: Each digestate was diluted with distilled water to a final volume of 125 ml, and 6 ml sodium chloride–hydroxylamine sulfate solution was added to reduce excess permanganate. After 30 seconds, 5 ml stannous sulfate solution was added and the bottle was immediately attached to the aeration apparatus. Mercury vapors were swept into a Plexiglass absorption cell and measured by a Coleman MAS-50 mercury analyzer system (*26*). Detection limit was 0.02 mg/kg.

DIGESTION AND ANALYSIS OF SAMPLES COLLECTED IN 1977

Mercury, Cadmium, and Lead—Mercury, cadmium, and lead residues in fish were oxidized by acid digestion in heated, enclosed glass bombs (45-ml acylation tubes, Regis Chemical Co.), which allow oxidation and recovery of all three meals with a single digestion. Digestion tubes were cleaned by successive rinsing with concentrated HNO_3, HCl, and ultrapure water (15–18 megohm-cm specific resistivity). Cleaned tubes were oven-dried, cooled, and covered with sheet polyethylene before the sample was introduced. Teflon caps for the tubes were soaked successively for several hours in boiling concentrated HNO_3 and HCl, followed by a final rinsing with ultrapure water.

Approximately 2 g ($\pm$0.01 g) thawed fish homogenate was weighed into tared digestion tubes. Sub-boiling-point, distilled concentrated HNO_3 (2 ml) and double-distilled $HClO_4$ (1 ml) were added and the mixture was vortex mixed, capped loosely, and allowed to predigest overnight at room temperature (*1*). The mixture was vortex mixed again, and the bomb was sealed and placed in a heated aluminum block (65°C) for 48 hours. The sample was quantitatively transferred and diluted to a final weight of 50 $\pm$ 0.01 g with 1 percent HCl. Diluted digestates were stored in cleaned polypropylene bottles (*38*) for cadmium and lead analyses or borosilicate test tubes for mercury analysis (*14*).

Mercury was determined by flameless atomic absorption spectrophotometry (AAS) (*28*). The analytical system was automated with a Technicon Autosampler IV and Proportioning Pump III, with appropriate pump tubes, pulse suppressors, mixing coils, and locally fabricated phase separator (Figure 2). Atomic absorption measurements were made on a Perkin-Elmer Model 305B spectrophotometer, using the 253.7-nm resonance line from an electrodeless discharge lamp. Scale expansion up to 10× was used when appropriate. The detection limit was about 0.01 mg/kg. The absorption cell was constructed from a Pyrex tube about 100 mm long and 6 mm I.D. with quartz end windows. Side arms of 4-mm-O.D. Pyrex were attached near each end of the cell for vapor passage. The cell was heated to 35–40°C with a high-intensity radiant heat projector (Cole-Parmer Dyna Lume Model 3151–6) to prevent condensation on the end windows.

Lead and cadmium were measured with a Perkin-Elmer Model 305B spectrophotometer equipped with an HGA–2100 graphite furnace and an AS–1 autosampling system. Table 2 specifies the instrument conditions for measuring each element. A four-point additions procedure to correct for chemical and matrix interferences was performed on each diluted digestate:

(1) Digestate + 0.00 ppm Pb; + 0.00 ppm Cd
(2) Digestate + 0.02 ppm Pb; + 0.002 ppm Cd
(3) Digestate + 0.04 ppm Pb; + 0.004 ppm Cd
(4) Digestate + 0.06 ppm Pb; + 0.006 ppm Cd

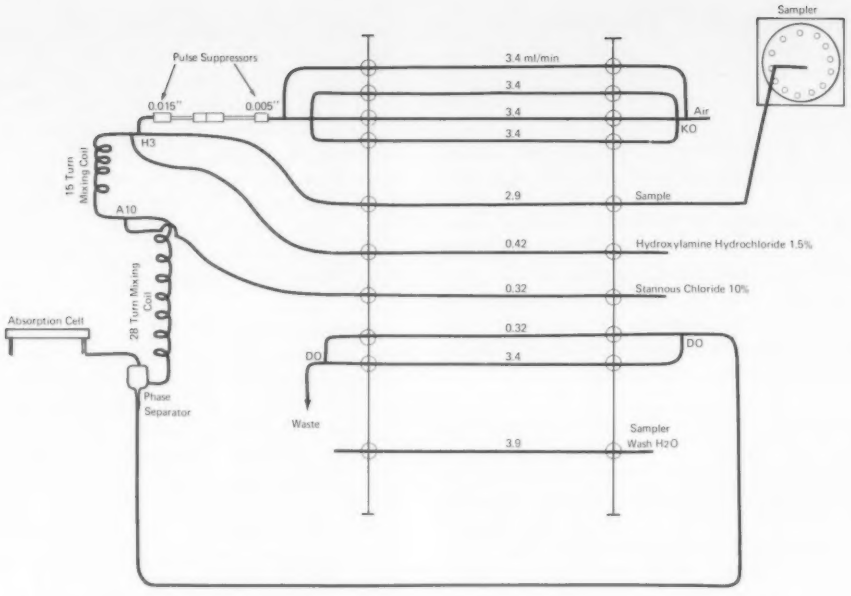

FIGURE 2. *Flow scheme for automated digestion and determination of mercury.*

TABLE 2. *Instrumental conditions for atomic absorption measurement of cadmium, lead, arsenic, and selenium in freshwater fish from the United States, 1977*

Condition	Cadmium	Lead	Arsenic	Selenium
Wavelength, nm	228.8	283.3	193.7	196.0
Spectral band width, nm	0.7	0.7	0.7	2.0
Temperature ramping	no	no	no	no
Drying time, °C (time, seconds)	90(30)	90(30)	—	—
Charring time, °C (time, seconds)	400(20)	400(20)	—	—
Atomization temp., °C (time, seconds)	2200(7)	2300(7)	—	—
Purge gas and flow (ml/minute)	Ar(20)	Ar(20)	Ar	Ar
Gas mode	normal	normal	EDL	—
Source	EDL	EDL	no	EDL
Background correction	D_2Arc	D_2Arc	5 mv	no
Scale expansion	3×	10×	PR I	3×
Recorder full scale	10 mv	10 mv	—	10 mv
AS-1 injection volume	10 µl	10 µl	none	—
Tube type	uncoated	uncoated	—	—
Quartz cell temp., °C	—	—	—	800
Reductant	—	—	1000	$NaBH_4$
Reaction flask volume, ml	—	—	$NaBH_4$	10
Analysis cycle time	—	—	10	PR II
Detection limit, mg/kg	0.01	0.1	0.05	0.05

The average of two injections was used as one additions point. Correlation coefficients (r) below 0.999 for the line of best fit were rejected, and corresponding digestates were rerun.

Arsenic and Selenium—For arsenic digestions, approximately 2 g (±0.01 g) thawed fish homogenate was weighed into a tared, 100-ml Kjeldahl flask. Sub-boiling-point, distilled concentrated HNO_3 (5 ml) was added, the flask was loosely covered with sheet polyethylene, and the mixture was predigested overnight at room temperature. Subboiled HNO_3 (25 ml) and double distilled $HClO_4$ (3 ml) were added, and the flask was heated on a micro-Kjeldahl rack to drive off HNO_3. After digestion had proceeded through the $HClO_4$ fuming and reaction stages, additional heat was applied to drive off the acid. When about 0.5 ml $HClO_4$ remained, the flasks were removed from the heat, cooled, and contents were diluted with 3 percent HCl to final volumes of 100 ml. Diluted digestates were stored in linear polyethylene bottles before analysis.

Predigestion sample preparation for selenium was the same as that for arsenic. Following predigestion, 25 ml subboiled HNO_3 and 1 ml double-distilled $HClO_4$ were added and the flask was heated to drive off HNO_3. Digestion was allowed to proceed through $HClO_4$ fuming and reaction stages. The $HClO_4$ reaction stage was characterized by an initial foaming with subsequent clearing (decoloration) of the digestate. Care was taken to terminate the digestion before $HClO_4$ was driven off (*88*). Cooled digestates were diluted to 100 ml with 3 percent HCl and transferred to linear polyethylene bottles.

Arsenic and selenium were analyzed with a Perkin-Elmer MHS–1 mercury-hydride system in conjunction with a Perkin-Elmer Model 305B spectrophotometer.

Instrument conditions for measuring the elements are listed in Table 2. On the MHS–1 system, a reaction flask containing 10 ml diluted digestate was installed on a polypropylene manifold. Argon was recycled in a closed circuit through the reaction flask and a heated quartz cell. An $NaBH_4$ pellet (Alfa Ventron) was dispensed into the flask and reacted with the digestate acid to form hydrogen, which reduced the metals to volatile arsine and hydrogen selenide. The gas stream flushed the hydrides into the heated quartz cell where they were decomposed and measured. Complete analysis, from addition of the digestate to readout on the spectrophotometer, proceeded automatically for each metal.

STATISTICAL TREATMENT

A two-way analysis of variance (ANOVA) with stations and years as main effects was used to test two null hypotheses: (1) There was no significant difference in metal residue concentrations due to location, and (2) there was no effect due to time (1972–73 vs. 1976–77). In general, stations were confounded by species differences across years. The following adjustments were made in the data sets before statistical testing:

(a) Absolute values were used for all less than (<) data.

(b) Selenium values for 1972 and 1973 were available for only 44 of the stations sampled in 1977. Therefore, the two-way ANOVA data set for selenium consisted of 44 stations and 3 years (1972, 1973, and 1977).

(c) The detection limit for cadmium in 1977 samples (0.01 ppm) was one-fifth the limit in 1972 and 1976 samples (0.05 ppm). Therefore, we adjusted all 1977 cadmium values <0.05 mg/kg to 0.05, to eliminate bias due to differences in detection limits. The two-way ANOVA data set consisted of 82 matching stations and two time periods (1972 vs. 1976–77).

(d) The two-way ANOVA data set for lead and mercury also consisted of 82 stations and two time periods (1972 vs. 1976–77).

(e) Detection limits for arsenic were 0.05 ppm for the 1972 and 1977 samples and 0.25 ppm for 1976 samples. Because most of the 1977 arsenic concentrations were <0.25 mg/kg, the results of arsenic residue analyses for the 1976 stations were not included in the two-way ANOVA set; consequently, only 44 matching stations and 2 years (1972 vs. 1977) remained.

An error sum of squares to estimate variation within individual samples was obtained from a preliminary two-way ANOVA with an unbalanced cell size (*52*). The method of weighted squares of means (*52*) was then used to make inferences about main effects. All analyses were performed on log-transformed data [$\log_{10}(1 + \text{conc.})$]

Results

The precision and accuracy of the 1976–77 trend-monitoring analyses were determined by duplicate sample sets and samples containing inorganic spikes. A duplicate sample set is defined as two aliquots of tissue from the same sample composite carried through the entire digestion-analytical procedure. The average difference of duplicate sets was within 25 percent for all elements, except for mercury in 1976 samples (37 percent) and lead in 1977 samples (70 percent). Average recoveries from spiked samples were within 10 percent of the added quantity, except for mercury in 1976, for which average recovery was 64 percent (Table 3a). Analyses of reference materials yielded average values within the specified certification ranges for all elements (Table 3b).

TABLE 3. *Quality control results of the 1976–77 trend-monitoring analyses*

a. Recoveries of elements from spiked samples

	1976				1977			
ELEMENT	*n*	CONCN., µG/ML	MEAN RECOVERY, %	S.D.	*n*	CONCN., µG/ML	MEAN RECOVERY, %	S.D.
Cadmium	14	0.1	92	10	7	0.004	106	7
Lead	13	0.1	91	10	8	0.040	102	12
Mercury	6	0.001	64	15	8	0.010	98	12
Arsenic	14	0.1	98	17	17	0.004	94	10
Selenium	—	—	—	—	15	0.004	101	17

b. Reference materials—1977

REFERENCE MATERIAL [1]	ELEMENT	CERTIFIED CONCN. RANGE, µG/G	*n*	MEAN CONCN., µG/G	S.D.
NBS Bovine liver	cadmium	0.27 ± 0.04	11	0.31	0.02
NBS Bovine liver	lead	0.34 ± 0.08	9	0.38	0.05
NBS Bovine liver	mercury	0.016 ± 0.002	12	0.016	0.005
FDA Cod	arsenic	11 ± 2	13	10.5	1.37
NBS Albacore tuna	arsenic	3.3 ± 0.4	18	3.27	0.35
FDA Cod	selenium	1.4 ± 0.4	13	1.36	0.21
NBS Albacore tuna	selenium	3.6 ± 0.4	18	3.68	0.40

NOTE: *n* = number of samples; S.D. = standard deviation; FDA = Food and Drug Administration; NBS = National Bureau of Standards.

Locations, species sampled, average size, and trace element concentrations for 1976–77 trend-monitoring samples are listed in Table 4. Eighty-three samples collected in 1976 were analyzed by the EPA Region VII Laboratory. The 1977 samples (157 samples of 163 collected) were analyzed by CNFRL. The data ranges in mg/kg wet weight were Pb, 0.10–4.92; Hg, 0.01–0.84; Cd, 0.01–1.04; As, 0.05–2.92; and Se, 0.05–

2.87. To examine potential temporal trends of trace element concentrations, 1976–77 data for lead, mercury, cadmium, and arsenic were compared with those reported by Walsh et al. (*87*) for the same stations in 1972. Because selenium concentrations were not reported for the 1976 collections, 1977 selenium concentrations were compared with those from the same stations in 1972 and 1973.

TABLE 4. *Concentrations of cadmium, lead, mercury, arsenic, and selenium in whole-fish samples collected for the National Pesticide Monitoring Program, 1976–77*

Station Number and Location (Figure 1)	Species [1]	No. of Fish	Average Size: Length, inches	Average Size: Weight, lb	Residues, mg/kg Wet Weight: Cadmium	Lead	Mercury	Arsenic	Selenium
ATLANTIC COASTAL STREAMS									
1. Pennobscot River, Old Town, Me.	white perch	5	9.8	0.5	<0.05	0.12	0.23	<0.25	—
	white sucker [2]	5	14.3	1.18	—[3]	—	—	—	—
		5	14.1	1.16	<0.05	0.13	0.20	<0.25	—
51. Kennebec River, Hinckley, Me.	yellow perch	5	9.6	0.36	<0.05	0.13	0.34	<0.25	—
	white sucker	5	14.2	1.22	0.06	0.13	0.16	<0.25	—
		5	14.5	1.22	—	—	—	—	—
52. Lake Champlain, Burlington, Vt.	northern pike	5	17.8	1.48	<0.05	<0.10	0.21	<0.25	—
	brown bullhead	5	11.1	0.8	<0.05	0.34	0.04	<0.25	—
		5	10.7	0.64	—	—	—	—	—
53. Merrimac River, Lowell, Mass.	largemouth bass	4	9.7	0.48	<0.05	0.21	—	<0.25	—
	white sucker	5	11.8	0.64	0.06	1.23	<0.02	<0.25	—
		5	12.6	0.76	—	—	—	—	—
2. Connecticut River, Windsor Locks, Conn.	white perch	5	9.2	0.46	0.11	0.40	0.27	<0.25	—
	white catfish	5	11.6	0.74	0.25	1.19	0.08	<0.25	—
		5	12.1	0.88	0.16	0.79	0.12	<0.25	—
3. Hudson River, Poughkeepsie, N.Y.	largemouth bass	5	12.1	0.92	<0.05	0.10	0.09	<0.25	—
	goldfish	5	11.3	1.18	0.20	3.07	0.05	<0.25	—
		5	11.1	1.08	0.32	3.83	0.06	0.68	—
54. Raritan River, Highland Park, N.J.	carp	5	17.3	3.14	<0.05	0.23	0.05	<0.25	—
4. Delaware River, Camden, N.J.	white perch	5	7.0	0.2	0.22	1.57	0.14	0.21	1.83
	white sucker	5	15.4	1.7	0.20	0.98	0.12	0.10	0.51
		5	15.2	1.6	0.16	1.17	0.14	0.10	0.54
5. Susquehanna River, Conowingo Dam, Md.	white perch	5	6.3	0.1	0.05	0.44	0.10	0.28	1.62
	channel catfish	6	10.7	0.4	0.04	0.19	0.05	<0.05	0.65
	carp	5	16.6	1.9	0.02	0.34	0.08	0.11	0.66
		5	16.7	1.9	0.05	0.26	0.11	0.11	0.52
6. Potomac River, Little Falls, Md.	largemouth bass	5	12.2	1.2	<0.01	0.15	0.20	<0.05	0.42
	carp	5	19.0	3.9	0.04	0.78	0.12	0.13	0.40
		5	17.5	2.9	0.04	0.33	0.09	0.16	0.49
55. James River, Richmond, Va.	smallmouth bass	5	9.2	0.4	0.03	0.14	0.12	0.06	0.49
	redhorse	4	16.0	1.8	0.81	0.32	0.25	0.10	0.21
		4	17.5	2.2	0.36	0.15	0.18	0.07	0.36
7. Roanoke River, Roanoke Rapids, N.C.	white catfish	5	11.1	0.54	<0.05	0.16	0.05	<0.25	—
		5	12.2	0.86	—	—	—	—	—
8. Cape Fear River, Elizabethtown, N.C.	gizzard shad	5	13.7	1.06	0.08	0.40	0.04	0.26	—
		5	13.3	0.96	—	—	—	—	—
56. Pee Dee River, Dongola, S.C.	white catfish	4	15.1	1.35	—	—	—	—	—
		5	14.7	1.26	<0.05	0.20	0.45	<0.25	—
9. Cooper River, Summerton, S.C.	striped bass	2	26.0	7.3	—	—	—	—	—
	channel catfish	5	13.3	0.86	<0.05	0.28	<0.02	<0.25	—
		5	13.9	0.8	—	—	—	—	—
10. Savannah River, Savannah, Ga.	largemouth bass	4	10.9	0.83	<0.05	<0.10	0.12	<0.25	—
	channel catfish	3	15.1	1.33	0.08	<0.10	0.06	0.63	—
	white catfish	3	13.8	1.63	0.17	<0.10	0.14	1.46	—
	carp	4	14.6	1.83	—	—	—	—	—
57. Altamaha River, Doctortown, Ga.	brown bullhead	5	14.6	1.56	—	—	—	—	—
	channel catfish	3	17.4	1.9	<0.05	0.30	0.14	<0.25	—
	largemouth bass	5	15.4	2.1	<0.05	0.18	0.34	<0.25	—
11. St. Johns River, Welaka, Fla.	inactive [4]								
12. St. Lucie Canal, Indiantown, Fla.	largemouth bass	5	15.3	2.26	<0.05	<0.10	<0.02	<0.25	—
	white catfish	5	13.4	1.24	<0.05	0.14	0.06	<0.50	—
	channel catfish	3	19.8	3.03	—	—	—	—	—

TABLE 4. (cont'd.). *Concentrations of cadmium, lead, mercury, arsenic, and selenium in whole fish samples collected for the National Pesticide Monitoring Program, 1976–77*

Station Number and Location (Figure 1)	Species [1]	No. of Fish	Average Size: Length, inches	Average Size: Weight, lb	Residues, mg/kg Wet Weight: Cadmium	Lead	Mercury	Arsenic	Selenium
101. Androscogin River, Lewiston, Me.	yellow perch	5	9.3	0.38	<0.05	0.16	0.20	<0.25	—
	white sucker	5	12.2	0.68	<0.05	0.24	0.11	<0.25	—
		5	12.4	0.38	—	—	—	—	—
GULF COAST STREAMS									
58. Suwanee River, Old Town, Fla.	inactive								
13. Apalachicola River, Jim Woodruff Dam, Ala.	largemouth bass	5	14.1	1.56	0.02	0.18	0.11	0.11	0.23
	spotted sucker	5	17.5	2.38	0.02	<0.10	0.11	<0.05	0.52
		5	18.4	2.82	0.03	0.16	0.17	0.07	0.52
59. Alabama River, Chrysler, Ala.	mixed species [5]	3	13.5	1.77	0.02	0.19	0.16	0.13	0.34
	largemouth bass	5	15.9	2.3	0.03	<0.10	0.55	0.20	0.31
	freshwater drum	8	10.2	0.66	0.03	0.15	0.07	0.21	0.65
14. Tombigbee River, McIntosh, Ala.	mixed species [6]	4	13.9	2.3	0.03	0.20	0.18	0.18	0.44
	freshwater drum	5	11.7	0.78	0.04	0.13	0.50	0.09	0.77
15. Mississippi River, Luling, La.	freshwater drum	5	13.0	1.1	0.03	0.17	0.03	0.26	0.44
		5	14.1	1.58	0.03	0.12	0.10	0.13	0.45
		5	14.3	1.58	0.02	0.17	0.12	0.18	0.67
60. Brazos River, Richmond, Tex.	alligator gar	3	18.5	1.0	0.02	<0.10	0.31	0.08	0.24
	striped mullet	5	14.1	1.3	0.03	0.48	0.01	1.48	0.40
	striped mullet [2]	5	13.3	1.1	0.02	0.46	0.02	1.40	0.24
	gizzard shad	5	9.9	0.4	0.01	0.65	0.04	0.60	0.26
61. Colorado River, Wharton, Tex.	largemouth bass	3	9.4	0.4	0.01	0.10	0.12	0.05	0.44
	channel catfish	5	12.4	0.6	0.06	0.14	0.03	<0.05	0.30
		5	13.3	0.8	0.04	<0.10	0.05	0.07	0.29
	gizzard shad	5	12.2	0.8	0.01	0.20	0.01	0.85	0.32
62. Nueces River, Mathis, Tex.	largemouth bass	5	13.3	1.5	<0.01	<0.10	0.18	0.21	0.29
	gizzard shad	5	11.3	0.6	0.03	0.61	0.02	0.53	0.29
		5	11.3	0.7	0.03	0.56	0.02	0.37	0.47
16. Rio Grande, Brownsville, Tex.	channel catfish	3	12.6	0.63	—	—	—	—	—
	gizzard shad	5	11.6	0.56	—	—	—	—	—
		5	11.8	0.52	—	—	—	—	—
63. Rio Grande, Elephant Butte, N.M.	not collected [7]								
64. Rio Grande, Alamosa, Colo.	brown trout	5	11.18	0.62	—	—	—	—	—
	white sucker	5	10.7	0.52	<0.05	0.39	0.03	<0.25	—
		5	10.8	0.52	<0.05	0.10	0.04	<0.25	
65. Pecos River, Red Bluff Lake, Tex.	not collected								
113. San Antonio River, McFaddin, Tex.	channel catfish	3	19.2	4.8	0.02	0.25	0.13	<0.05	0.41
	alligator gar	3	22.9	2.2	0.17	0.11	0.28	0.13	0.17
	smallmouth bass	3	18.3	6.6	0.02	0.71	0.06	0.09	0.66
GREAT LAKES DRAINAGE									
17. Genessee River, Scottsville, N.Y.	pumpkinseed	5	5.1	0.2	0.03	0.62	0.09	0.06	0.28
	carp	5	15.9	2.3	0.02	0.35	0.04	0.09	0.19
		5	15.5	2.2	0.03	0.23	0.04	0.14	0.37
66. St. Lawrence River, Massena, N.Y.	smallmouth bass	5	12.6	1.2	0.01	0.14	0.26	0.67	0.45
	white sucker	5	14.1	1.3	0.03	0.41	0.19	0.16	0.31
18. Lake Ontario, Port Ontario, N.Y.	rock bass	5	8.6	0.5	0.03	0.20	0.36	0.31	0.36
	channel catfish	5	14.1	0.9	0.02	0.22	0.07	0.10	0.30
		5	14.2	0.9	0.04	0.20	0.07	0.16	0.29
19. Lake Erie, Erie, Pa.	yellow perch	5	8.6	0.4	0.03	<0.10	0.08	0.11	0.67
	white sucker	5	11.3	0.6	0.05	0.19	0.05	0.17	0.69
		5	11.4	0.7	0.05	0.16	0.05	0.17	0.50
20. Lake Huron, Bay Port, Mich.	carp	5	18.7	3.72	0.04	0.49	0.23	0.10	0.35
	carp [2]	5	17.1	2.9	0.02	0.22	0.05	0.13	0.64
	yellow perch	5	7.6	0.2	0.02	0.23	0.04	<0.05	0.44
		5	7.5	0.2	0.01	0.23	0.03	<0.05	0.34

TABLE 4. (cont'd.). *Concentrations of cadmium, lead, mercury, arsenic, and selenium in whole fish samples collected for the National Pesticide Monitoring Program, 1976–77*

Station Number and Location (Figure 1)	Species [1]	No. of Fish	Average Size: Length, inches	Average Size: Weight, lb	Residues, mg/kg Wet Weight: Cadmium	Lead	Mercury	Arsenic	Selenium
21. Lake Michigan, Sheboygan, Wis.	bloater	5	10.1	0.52	0.01	0.15	0.03	2.92	0.30
		5	10.0	0.56	0.02	0.15	0.04	2.91	0.44
	lake trout	5	27.5	6.6	<0.01	<0.10	0.19	1.20	0.60
		4	23.8	4.5	0.02	<0.10	0.19	1.33	0.46
22. Lake Superior, Bayfield, Wis.	lake trout	6	24.7	5.0	0.02	<0.10	0.43	0.56	0.32
	lake whitefish	3	20.3	2.86	0.05	<0.10	0.03	0.47	0.57
102. Lake Superior, Keeweenaw Point, Mich.	bloater	5	10.4	0.60	0.03	0.12	0.11	1.33	0.75
		5	10.3	0.60	0.30	<0.10	0.10	1.19	0.73
	lake trout	4	22.0	4.40	0.02	<0.10	0.10	0.39	0.52
103. Lake Superior, Whitefish Point, Mich.	lake trout	4	23.9	5.1	0.02	0.14	0.26	0.43	0.52
	lake whitefish	5	19.2	3.0	0.04	0.29	0.05	0.41	0.61
		5	19.8	2.8	0.04	0.11	0.07	0.97	0.57
104. Lake Michigan, Beaver Island, Mich.	bloater	4	11.3	0.6	0.02	0.12	0.04	2.41	0.44
	lake trout	5	25.7	5.9	<0.01	<0.10	0.27	0.92	0.52
105. Lake Michigan, Saugatuck, Mich.	bloater	5	11.4	0.6	—	—	—	—	—
		5	11.3	0.6	0.03	0.31	0.05	2.30	0.41
	lake trout	5	24.7	4.7	0.01	<0.10	0.25	0.94	0.71
MISSISSIPPI RIVER SYSTEM									
67. Allegheny River, Natrona, Pa.	walleye	3	12.4	0.63	—	—	—	—	—
	silver redhorse	5	10.6	0.48	—	—	—	—	—
	golden redhorse	5	13.2	0.94	0.07	0.19	0.07	0.26	—
23. Kanawha River, Winfield, W.Va.	black crappie	5	6.3	0.14	—	—	—	—	—
	carp	5	14.8	1.44	<0.05	<0.10	<0.02	<0.25	—
	brown bullhead	5	9.6	0.46	—	—	—	—	—
68. Wabash River, New Harmony, Ind.	largemouth bass	5	10.7	0.72	0.06	0.17	—	<0.25	—
	carp	5	16.8	2.7	—	—	—	—	—
		5	16.1	2.38	0.18	0.22	0.10	<0.25	—
24. Ohio River, Marietta, Ohio	sauger	5	13.9	1.18	<0.05	0.26	0.10	<0.25	—
	carp	4	17.2	2.92	0.22	2.49	0.05	<0.25	—
69. Ohio River, Cincinnati, Ohio	sauger	5	13.7	1.04	<0.05	0.10	0.10	<0.25	—
	channel catfish	5	11.1	0.5	—	—	—	—	—
		4	15.5	1.3	<0.05	0.42	0.04	<0.25	—
	smallmouth buffalo	5	12.2	1.08	—	—	—	—	—
		3	13.7	1.8	—	—	—	—	—
70. Ohio River, Metropolis, Ill.	largemouth bass	5	14.2	1.72	<0.05	<0.10	0.30	<0.25	—
	carp	5	16.5	2.52	—	—	—	—	—
		5	16.2	2.12	0.18	0.25	0.15	<0.25	—
25. Cumberland River, Clarksville, Tenn.	largemouth bass	4	9.8	0.52	—	—	—	—	—
	carp	5	13.4	1.16	—	—	—	—	—
	spotted sucker	4	12.2	0.76	—	—	—	—	—
71. Tennessee River, Savannah, Tenn.	not collected								
106. Lake Huron, Alpena, Mich.	lake trout	5	23.4	4.3	<0.01	<0.10	0.23	0.51	0.71
	white sucker	5	19.0	2.8	0.04	0.39	0.07	0.11	0.66
		6	16.3	1.9	0.02	0.28	0.03	0.10	0.87
		5	12.7	0.9	0.02	0.13	0.02	0.16	0.55
107. Lake St. Clair, Mt. Clemens, Mich.	walleye	5	20.3	2.7	0.06	0.13	0.84	0.24	0.83
	carp	5	20.4	4.5	0.15	1.83	0.60	0.13	0.86
		5	20.1	4.5	0.16	0.78	0.32	0.09	0.76
108. Lake Erie, Port Clinton, Ohio	walleye	5	15.9	1.6	<0.01	<0.10	0.09	0.36	0.26
	carp	5	15.9	2.5	0.03	0.19	0.03	0.15	0.44
		5	16.7	2.4	0.03	0.21	0.04	0.21	0.52
109. Lake Ontario, Roosevelt Beach, N.Y.	inactive								
110. Lake Ontario, Cape Vincent, N.Y.	inactive								
72. Wisconsin River, Woodsman, Wis.	smallmouth bass	5	12.9	1.12	—	—	—	—	—
	carp	5	17.2	2.3	0.07	0.23	0.14	<0.25	—
		5	17.7	2.46	—	—	—	—	—

TABLE 4. (cont'd.). *Concentrations of cadmium, lead, mercury, arsenic, and selenium in whole fish samples collected for the National Pesticide Monitoring Program, 1976–77*

Station Number and Location (Figure 1)	Species [1]	No. of Fish	Average Size		Residues, mg/kg Wet Weight				
			Length, inches	Weight, lb	Cadmium	Lead	Mercury	Arsenic	Selenium
73. Des Moines River, Keosauqua, Iowa	carp	5	15.6	1.94	0.06	0.27	0.07	<0.25	—
		5	15.4	1.94	1.04	0.41	—	<0.25	—
	walleye	3	16.5	1.53	<0.05	<0.10	0.06	<0.25	—
26. Illinois River, Beardstown, Ill.	black crappie	5	8.2	0.32	<0.05	0.23	0.09	<0.25	—
	carp	5	14.1	1.22	<0.05	0.32	0.05	<0.25	—
		5	14.2	1.40	—	—	—	—	—
74. Mississippi River, Little Falls, Minn.	walleye	5	11.1	0.5	—	—	—	—	—
	black bullhead	5	8.2	0.32	0.06	0.10	0.32	<0.25	—
		5	7.9	0.28	—	—	—	—	—
27. Mississippi River, Guttenberg, Iowa	largemouth bass	5	14.1	1.72	<0.05	0.55	0.05	<0.25	—
	carp	5	19.9	3.56	<0.05	0.30	0.05	<0.25	—
		5	21.3	4.48	—	—	—	—	—
75. Mississippi River, Cape Girardeau, Mo.	white crappie	5	8.7	0.24	<0.05	0.13	0.17	<0.25	—
	carp	5	14.4	1.26	0.45	0.49	<0.02	0.30	—
		5	15.4	1.68	—	—	—	—	—
76. Mississippi River, Memphis, Tenn.	smallmouth buffalo	3	17.3	2.63	0.02	<0.10	0.04	0.14	0.25
		3	16.4	2.97	0.03	0.13	0.05	0.11	0.33
28. Arkansas River, Pine Bluff, Ark.	bluegill	5	6.3	0.1	0.02	0.27	0.05	0.12	0.23
	carp	5	19.8	3.78	0.05	0.18	0.07	0.12	0.33
29. Arkansas River, Keystone Reservoir, Okla.	largemouth bass	5	13.7	1.8	<0.01	<0.10	0.17	0.20	0.67
	carp	5	17.9	2.1	0.12	0.39	0.15	<0.05	0.54
		5	17.8	2.4	0.09	0.44	0.21	0.07	0.65
77. Arkansas River, John Martin Reservoir, Colo.	inactive								
78. Verdigris River, Oologah, Okla.	white bass	5	12.4	0.9	0.02	0.11	0.07	0.55	0.60
	carp	5	17.8	2.5	0.54	0.75	0.09	0.08	0.50
		5	16.1	2.1	0.78	0.82	0.09	0.09	0.44
79. Canadian River, Eufaula, Okla.	white bass	5	13.9	1.4	<0.01	<0.10	0.34	0.65	0.35
	river carpsucker	5	13.0	1.1	0.02	0.38	0.13	0.08	0.28
		5	12.2	0.7	0.02	0.38	0.06	0.16	0.38
30. White River, DeValls Bluff, Ark.	white crappie	5	8.5	0.2	0.02	0.15	0.18	0.12	0.24
	bigmouth buffalo	3	15.1	1.87	0.04	0.17	0.13	0.06	0.54
		3	15.5	1.87	0.04	0.24	0.14	0.12	0.41
80. Yazoo River, Redwood, Miss.	black crappie	5	9.3	0.9	<0.01	0.10	0.14	0.16	0.43
	bigmouth buffalo	5	15.0	2.0	0.02	0.10	0.04	0.10	0.41
		5	15.5	2.2	0.04	0.11	0.06	0.09	0.38
81. Red River, Alexandria, La.	smallmouth buffalo	5	17.1	2.8	0.01	0.19	0.10	<0.05	0.22
		5	17.3	3.14	0.02	0.12	0.13	0.05	0.32
		5	17.0	2.94	0.01	0.15	0.10	<0.05	0.35
82. Red River, Lake Texoma, Okla.	largemouth bass	5	10.4	0.6	0.03	0.31	0.12	0.11	0.72
	gizzard shad	5	10.4	0.4	0.05	1.14	0.02	0.55	0.89
		5	9.9	0.3	0.05	1.44	0.02	0.61	0.86
83. Missouri River, Hermann, Mo.	freshwater drum	5	16.3	2.2	0.03	<0.10	0.24	0.06	0.66
31. Missouri River, Nebraska City, Neb.	goldeye	5	12.5	0.7	0.03	2.43	0.09	<0.05	0.73
	carp	5	14.8	1.7	0.06	0.75	0.03	0.10	1.28
		5	15.3	1.8	0.06	0.86	0.05	0 13	1.52
32. Missouri River, Garrison Dam, N.D.	walleye	5	17.3	1.64	<0.01	0.10	0.22	0.18	0.70
	white sucker	5	14.2	1.18	0.03	0.47	0.08	0.32	0.73
		5	14.3	1.18	0.03	0.30	0.07	0.23	0.73
33. Missouri River, Great Falls, Mont.	goldeye	9	12.7	0.7	0.06	0.13	0.12	0.08	1.36
	longnose sucker	3	16.6	1.97	0.18	0.32	0.06	0.42	0.33
	redhorse sucker	5	17.1	2.36	0.44	0.14	0.15	0.10	0.29
84. Big Horn River, Hardin, Mont.	goldeye	10	12.4	0.5	0.03	0.11	0.19	0.06	2.87
	white sucker	10	14.2	1.2	0.01	0.10	0.09	0.09	1.71
		10	12.5	0.8	<0.01	<0.10	0.05	0.06	1.07
85. Yellowstone River, Sidney, Mont.	sauger	5	12.7	0.58	0.02	<0.10	0.22	0.06	1.75
	carp	5	14.8	1.62	0.04	<0.10	0.12	0.16	0.88
		5	20.7	3.82	0.24	0.17	0.15	0.08	0.82
86. James River, Olivet, S.D.	goldeye	5	14.1	0.9	0.03	0.92	0.23	<0.05	0.82
	carp	5	16.1	2.0	0.05	1.95	0.07	0.06	0.53
		5	16.2	2.0	0.10	1.08	0.09	0.07	0.57
87. North Platte River, Lake McConaughy, Neb.	walleye	5	17.2	1.6	<0.01	0.42	0.04	0.81	0.73
	carp	5	19.1	3.0	0.09	0.54	0.17	0.13	0.81
		5	20.1	3.8	0.08	0.94	0.10	0.20	0.63
88. South Platte River, Brule, Neb.	green sunfish	5	5.4	0.1	0.01	1.25	0.06	0.09	2.08
	carp	5	10.1	0.5	0.02	0.49	0.01	0.14	2.05
		5	9.6	0.4	0.02	0.53	0.02	0.05	2.53

TABLE 4. (cont'd.). *Concentrations of cadmium, lead, mercury, arsenic, and selenium in whole fish samples collected for the National Pesticide Monitoring Program, 1976–77*

Station Number and Location (Figure 1)	Species [1]	No. of Fish	Average Size: Length, inches	Average Size: Weight, lb	Residues, mg/kg Wet Weight: Cadmium	Lead	Mercury	Arsenic	Selenium
89. Platte River, Louisville, Neb.	goldeye	5	12.8	0.7	0.03	0.97	0.10	0.07	1.02
	river carpsucker	5	13.4	1.0	0.06	1.33	0.11	0.13	1.02
		5	14.6	1.4	0.10	1.74	0.12	0.07	1.13
90. Kansas River, Bonner Springs, Kans.	freshwater drum	5	9.3	0.34	0.02	<0.10	0.10	0.08	0.83
	carp	5	19.1	3.5	0.07	0.16	0.13	<0.05	0.88
		5	19.3	3.58	<0.01	0.21	0.12	0.09	1.03
111. Mississippi River, Lake City, Minn.	walleye	5	21.1	3.76	0.06	0.25	0.11	<0.25	—
	white sucker	5	17.8	2.56	0.05	0.25	<0.02	<0.25	—
112. Mississippi River, Dubuque, Iowa	largemouth bass	5	9.6	0.48	<0.05	<0.10	0.05	<0.25	—
	carp	5	18.5	2.98	<0.05	0.14	0.04	<0.25	—
		5	17.9	2.84	—	—	—	—	—
116. Souris River, International Border, N.D.	northern pike	5	19.5	1.66	<0.05	0.37	—	<0.25	—
HUDSON BAY DRAINAGE									
34. Red River, Noyes, Minn.	sauger	5	13.1	0.62	<0.05	<0.10	0.45	<0.25	—
		5	12.4	0.52	—	—	—	—	—
	white sucker	5	13.9	1.14	0.16	<0.10	0.13	<0.25	—
COLORADO RIVER SYSTEM									
35. Green River, Vernal, Utah	channel catfish	5	12.5	0.54	—	—	—	—	—
	carp	5	13.6	1.3	—	—	—	—	—
		5	12.5	1.24	—	—	—	—	—
36. Colorado River, Imperial Reservoir, Ariz	channel catfish	5	16.8	1.34	<0.05	<0.10	<0.02	<0.25	—
	carp	Not collected			—	—	—	—	—
		5	14.8	1.84	<0.05	0.18	<0.02	0.30	—
91. Colorado River, Lake Havasu, Ariz.	channel catfish	4	19.0	2.28	—	—	—	—	—
	carp	4	15.9	1.88	—	—	—	—	—
92. Colorado River, Lake Mead, Nebr.	inactive								
93. Colorado River, Lake Powell, Ariz.	largemouth bass	5	13.8	1.46	<0.05	0.27	0.14	<0.25	—
	carp	5	13.5	1.1	0.21	0.35	0.15	0.26	—
		5	13.5	1.2	0.22	0.16	0.07	<0.25	—
94. Gila River, San Carlos Reservoir, Ariz.	not collected								
114. Bear Creek, Brigham City, Utah	channel catfish	5	13.3	0.9	—	—	—	—	—
	carp	5	11.5	0.96	—	—	—	—	—
		5	14.7	1.68	0.09	<0.10	—	<0.25	—
115. Colorado River, Yuma, Colo.	mullet	5	13.5	0.99	—	—	—	—	—
	carp	4	11.7	0.81	<0.05	0.23	<0.02	<0.25	—
	largemouth bass	5	12.0	1.02	<0.05	0.20	0.03	<0.25	—
INTERIOR BASINS									
37. Truckee River, Fernley, Nebr.	carp	5	16.1	3.02	0.03	0.26	0.37	0.07	0.19
	Tahoe sucker	5	8.8	0.34	0.02	0.14	0.39	0.15	0.20
		5	8.7	0.32	0.02	0.28	0.32	0.19	0.19
38. Utah Lake, Provo, Utah	walleye	5	19.8	3.62	—	—	—	—	—
	carp	5	18.9	3.32	—	—	—	—	—
		5	18.8	3.24	—	—	—	—	—
95. Bear River, Preston, Idaho	inactive								
CALIFORNIA STREAMS									
39. Sacramento River, Sacramento, Calif.	largemouth bass	5	12.5	1.2	0.02	<0.10	0.14	0.19	0.36
	goldfish	4	12.4	1.3	0.11	0.19	0.19	0.23	0.24
		3	10.2	0.7	0.06	0.21	0.09	0.22	0.40
40. San Joaquin River, Los Banos, Calif.	striped bass	5	13.0	0.9	<0.01	<0.10	0.08	0.33	1.67
	Sacramento blackfish	5	12.3	0.8	<0.01	<0.10	0.07	0.12	1.53
		5	11.0	0.6	<0.01	0.12	0.16	0.09	1.07

TABLE 4. (cont'd.). *Concentrations of cadmium, lead, mercury, arsenic, and selenium in whole fish samples collected for the National Pesticide Monitoring Program, 1976–77*

Station Number and Location (Figure 1)	Species [1]	No. of Fish	Average Size: Length, inches	Average Size: Weight, lb	Residues, mg/kg Wet Weight: Cadmium	Lead	Mercury	Arsenic	Selenium
COLUMBIA RIVER SYSTEM									
41. Snake River, Hagerman, Idaho	northern squawfish	5	15.3	1.18	—	—	—	—	—
	largescale sucker	5	13.7	0.92	—	—	—	—	—
		5	13.5	0.94	—	—	—	—	—
	rainbow trout	5	13.8	1.0	—	—	—	—	—
42. Snake River, Lewiston, Idaho	northern squawfish	5	15.2	1.14	<0.05	0.10	0.31	<0.25	—
	largescale sucker	5	15.4	1.28	0.10	0.12	0.07	0.50	—
		5	15.9	1.36	0.07	0.10	0.11	1.11	
43. Salmon River, Riggins, Idaho	largescale sucker	5	18.2	1.7	0.12	0.23	0.19	0.50	—
		4	17.9	2.1	—	—	—	—	—
96. Snake River, Ice Harbor, Wash.	channel catfish	5	13.0	0.54	0.10	0.21	0.10	0.61	—
	largescale sucker	5	14.7	1.14	0.09	0.17	0.04	<0.25	—
		5	14.8	1.12	0.07	0.11	0.05	<0.25	—
44. Yakima River, Granger, Wash.	black crappie	4	6.0	0.1	0.14	0.19	0.05	0.50	—
	largescale sucker	5	11.6	0.56	—	—	—	—	—
		5	12.1	0.66	<0.05	0.10	0.09	0.61	—
45. Willamette River, Oregon City, Oreg.	smallmouth bass	3	9.2	0.5	<0.05	0.12	0.13	<0.25	—
	chiselmouth	5	10.2	0.4	0.20	0.85	<0.02	1.15	—
		5	10.8	0.5	—	—	—	—	—
46. Columbia River, Booneville Dam, Oreg.	northern squawfish	5	13.5	0.9	<0.05	<0.10	0.23	<0.25	—
	largescale sucker	5	14.3	1.08	—	—	—	—	—
		5	12.4	0.82	0.15	<0.10	0.05	0.87	—
97. Columbia River, Pasco, Wash.	carp	5	11.4	0.66	<0.05	0.34	<0.02	<0.25	—
		5	11.6	0.7	<0.05	0.13	<0.02	0.35	—
117. Flathead River, Creston, Mont.	northern squawfish	5	13.6	0.74	—	—	—	—	—
	longnose sucker	4	15.2	1.1	—	—	—	—	—
		3	12.8	1.0	—	—	—	—	—
98. Columbia River, Grand Coulee, Wash.	yellow perch	5	7.9	0.26	<0.05	0.16	0.03	<0.25	—
	largescale sucker	5	16.4	1.58	—	—	—	—	—
		5	17.3	1.74	0.33	2.57	<0.02	0.30	—
PACIFIC COAST STREAMS									
47. Klamath River, Hornbrook, Calif.	yellow perch	5	9.2	0.4	<0.01	<0.10	0.09	<0.05	0.16
	Klamath sucker	5	12.4	0.7	<0.01	<0.10	0.06	0.11	<0 05
		5	12.5	0.7	0.01	0.10	0.03	0.12	0.08
48. Rogue River, Gold Ray Dam, Oreg.	black crappie	5	9.1	0.76	<0.01	<0.10	0.16	<0.05	0.11
	Klamath sucker	5	8.7	0.28	0.01	<0.10	0.02	0.11	0.06
		5	9.1	0.32	0.01	<0.10	0.03	0.13	0.12
ALASKAN STREAMS									
49. Chena River, Fairbanks, Alaska	Artic grayling	5	10.5	0.38	0.02	0.27	0.07	0.05	0.80
	longnose sucker	5	14.6	1.26	0.03	0.13	0.08	0.17	0.32
50. Kenai River, Soldatna, Alaska	not collected								
HAWAIIAN STREAMS									
99. Waikele Stream, Waipahu, Hawaii	Cuban limia	28	1.9	<0.10	0.05	0.38	0.04	0.08	0.77
		28	2.4	<0.10	0.05	0.51	0.05	0.10	0.80
100. Manoa Stream, Honolulu, Hawaii	Chinese catfish	3	6.9	0.2	0.11	0.80	0.12	0.10	0.43
	tilapia	33	6.9	0.23	0.03	2.28	0.02	0.27	0.38
		3	6.3	0.2	—	—	—	—	—
	Cuban limia	36	2.7	<0.10	—	—	—	—	—
		37	2.7	<0.10	0.03	4.93	0.05	0.49	0.12

[1] Common names for species from the continental United States follow those designated in "A list of common and scientific names of fishes from the United States and Canada", American Fisheries Society Special Publication No. 6, 3rd Edition, 1970.
[2] Where two or more rows of data follow a species name, the data represent replicate samples.
[3] Dashes (—) = not analyzed (sample not submitted, or inadequate digestion procedure, e.g., 1976 Se data).
[4] Inactive = stations so designated have been temporarily deleted fromverse conditions, etc.).
[5] Mixed species = white catfish, carp, and spotted sucker.
[6] Mixed species = freshwater drum, spotted sucker, and smallmouth buffalo.
[7] Not collected = personnel were unable to obtain fish samples (e.g., ad the NPMP collection station network.

As expected, location effects were highly significant ($P < 0.001$) for all five elements (Table 5). Effects due to time were significant for mercury, arsenic, selenium ($P < 0.001$), and cadmium ($P < 0.05$), but not for lead ($P = 0.213$). Interactions between main effects were not significant.

TABLE 5. *Results of two-way analysis of variance (weighted squares of means) on concentrations of lead, mercury, cadmium, arsenic, and selenium in freshwater fish, United States, 1976–77*

Source	Degrees of Freedom	Sum of Squares	Mean Square	*F*	Significance Level
Lead					
Stations	81	4.536	0.056	5.905	0.001
Years	1	0.014	0.014	1.553	0.213
Interaction	81	0.661	0.008	0.860	0.791
Error	368	3.490	0.009		
Mercury					
Stations	81	0.367	0.004	3.305	0.001
Years	1	0.023	0.023	16.874	0.001
Interaction	81	0.059	0.0007	0.531	0.999
Error	364	0.499	0.001		
Cadmium					
Stations	81	0.432	0.005	2.196	0.001
Years	1	0.015	0.015	6.434	0.011
Interaction	81	0.126	0.001	0.645	0.991
Error	368	0.894	0.002		
Arsenic					
Stations	43	0.804	0.018	7.875	0.001
Years	1	0.051	0.051	21.774	0.001
Interaction	43	0.090	0.002	0.887	0.671
Error	217	0.515	0.002		
Selenium					
Stations	43	2.579	0.60	13.173	0.001
Years	2	0.160	0.080	17.663	0.001
Interaction	86	0.273	0.003	0.698	0.976
Error	352	1.603	0.004		

Significant differences in mercury ($P < 0.001$) and cadmium concentrations ($P < 0.05$) were observed from 1972 to 1976–77 (Table 6); lead concentrations did not decline significantly, but arsenic concentrations increased significantly ($P < 0.001$). The two-way ANOVA indicated an effect for selenium due to time ($P < 0.001$, Table 6). Mean selenium concentrations for 1973 were significantly lower than those for 1972 selenium values (a priori test, $0.001 < P < 0.005$). A posteriori comparison (*54*) of mean selenium levels indicated no significant differences between 1972 and 1977 samples. Because of changes in laboratories, analytical procedures, station, fish species, size and age of fish, etc., these results should be used cautiously as temporal trend information.

Discussion

The average percent difference between duplicates for cadmium, mercury, arsenic, and selenium was considered acceptable, particularly because some duplicates had elemental concentrations near the detection limit. Recoveries from spiked samples, and reference materials, indicated satisfactorily accurate recovery of all elements in all years, except for mercury in the spiked samples for 1976. The low recovery (64 percent) of mercury (Table 3) suggests problems during sample digestion and analysis.

TABLE 6. *Mean concentrations of lead, mercury, cadmium, arsenic, and selenium in whole fish* [1]

Element	Mean Concentrations, mg/kg wet weight [2]		Significance (*P*)
	1972	1976–77	
Lead	0.354 (0.322–0.387)	0.338 (0.297–0.380)	>0.05 (NS)
Mercury	0.153 (0.142–0.164)	0.112 (0.099–0.126)	<0.001
Cadmium	0.112 (0.098–0.126)	0.085 (0.068–0.102)	<0.05
Arsenic	0.127 (0.108–0.146)	0.207 (0.184–0.231)	<0.001
Selenium [3]	0.604 (0.567–0.641)	0.576 (0.534–0.619)	<0.001

[1] Matching stations in years.
[2] Figures in parentheses are 95 percent confidence intervals.
[3] The mean concentration (and 95% CI) for selenium in 1973 was 0.455 (0.422–0.488) mg/kg. This was the only element having matching station data in 1973. The *P* value (<0.001) results from the two-way ANOVA on all 3 years.

Imprecision in lead-quality control data indicates that measured lead values may not accurately reflect average levels in fish tissues. The accuracy and precision of lead spike recoveries (91 ± 10 percent and 102 ± 12 percent) and National Bureau of Standards reference material analyses (Table 3) indicate that digestion and analytical techniques were satisfactory. Nonuniform distribution of lead in fish tissue and authors' inability to achieve complete homogenization were more likely the causes of imprecision. For example, in tuna, the lead content of epidermis tissue is several thousand times that in muscle tissue from the same fish (*9, 59*). Most of the lead in the epidermis is associated with mucus (*9*). Similarly, rainbow trout (*Salmo gairdneri*) accumulate lead in mucus and scales (*85, 86*). As a result, lead digestate concentrations are difficult to correlate with muscle tissue dry weights because of mucosal slime contamination (*89*).

To approximate normal background ranges for whole-fish trace element concentrations, station means of log-transformed individual data values (Table 4) were calculated for each element and arranged in order of increasing concentration. The stations with transformed mean values exceeding the 85th percentile were thus identified (Table 7). The antilog of this 85th percentile was arbitrarily used to distinguish stations with high metal concentrations. Though the 85th percentile may not be meaningful biologically, it was considered above background and potentially worth further study. Authors

TABLE 7. *Stations from which fish had trace element concentration equal to or exceeding the 85th percentile for the 1976–77 trend-monitoring collection*

Element	85th Percentile, mg/kg	Stations (In order of increasing concentration)
Cadmium	0.11	70, 68, 43, 107, 45, 24, 93, 2, 98, 3, 4, 33, 75, 73, 55, 78
Lead	0.44	99, 78, 87, 53, 88, 2, 107, 82, 98, 24, 4, 31, 86, 89, 3, 100
Mercury	0.19	43, 22, 1, 70, 66, 57, 83, 59, 51, 34, 74, 14, 37, 56, 107
Arsenic	0.38	3, 66, 82, 43, 22, 46, 44, 42, 103, 45, 10, 60, 102, 105, 104, 21
Selenium	0.82	82, 4, 90, 89, 85, 31, 40, 84, 88

attempted to suggest sources that could account for the higher values encountered at these stations. It should be recognized that relating specific sources of metals to elevated concentrations of the elements in freshwater fish is speculative. The intent is to provide a metal source perspective for the drainage basins to help clarify elevated metal concentrations in fish from these areas.

CADMIUM

Cadmium concentrations in freshwater fish had a range of 0.01–1.04 ppm, a mean of 0.067 ppm, and an 85th percentile of 0.11 ppm. The decrease in cadmium concentrations in fish since 1972 (Table 6) parallels cadmium metal production and consumption, which declined over the same period (*63*). NPMP stations at which fish had cadmium concentrations equal to or exceeding the 85th percentile (Table 7) are discussed below in relation to possible contaminant sources.

Atlantic Coastal Streams—Fish from four rivers, the Connecticut (station 2), the Hudson (station 3), the Delaware (station 4), and the James (station 55), all contained mean cadmium concentrations exceeding 0.17 ppm. Each of these river systems lies in a heavily industrialized area. A zinc smelting company, which is a primary cadmium producer, is on the Lehigh River, a tributary of the Delaware. Metal fumes from the smelter's low stacks have killed trees in the Lehigh Valley, and river sediments contained 5.4 ppm cadmium (*78*). The James River receives effluents from numerous chemical, fertilizer, and other industries (*16*).

Great Lakes Drainage—Like the Atlantic coastal streams, Lake St. Clair (station 107) is surrounded by a heavily industrialized area. Cadmium residues in fish from the area ranged from 0.06 to 0.16 ppm, apparently originating from a number of industrial sources in and near Detroit.

Mississippi River System—The 0.22-ppm cadmium level in carp from station 24 at Marietta, Ohio, may result from zinc-smelting activity from a primary cadmium producer located on the Ohio River at Monaco, Pennsylvania. Similarly, a lead-smelting and refining complex at East Helena, Montana may be the primary source of cadmium in fish collected at station 33 (Great Falls, Montana. Miesch and Huffman (*76*) found cadmium contamination in soil 10 miles from a smelter in Helena Valley. It was estimated that 290 tons of cadmium had been added to the soil within a radius of 1–19 km from the smelter stacks. Superphosphate fertilizers are a suspect source at station 73 (Des Moines River), where high cadmium levels may reflect substantial agricultural runoff. High cadmium fluxes have been reported for the Mississippi River as it flows through mineralized areas in Tennessee, Missouri, and Kentucky (*15*). Cadmium levels in carp at Cape Girardeau, Missouri (station 75, Mississippi River), are probably the result of numerous sources. A zinc company at Sauget, Illinois (East St. Louis), has discharged waste on the Mississippi floodplain, forming a black sludge containing 0.1 percent cadmium (*78*). A large lead-smelting facility at Herculaneum, Missouri, has discharged effluent directly into the river. The smelter's slag piles along the banks of the Mississippi contained from 19 to 250 ppm cadmium, and much slag has been bulldozed into the river (*78*). In addition, industrial and municipal sewage effluents from St. Louis and phosphate fertilizer runoff may contribute to the cadmium load at this station.

The highest cadmium mean value was in carp from station 78 on the Verdigris River at Oologah, Oklahoma. A zinc company in Bartlesville, Oklahoma, may supply cadmium to the upper Verdigris River area as a result of particulate fallout from smelter stack emissions (*55*). The 85th percentile value of 0.11 ppm was characteristic of fish from station 70 on the Ohio River and probably reflects the relatively heavy industrialization of the lower river area.

Colorado River System—Southwestern Colorado and north-central Utah contain major deposits of lead and zinc ores, and numerous active mines are located there (*61*). A geologic source of cadmium, as well as mine waste drainage into tributaries of the Colorado River, could account for elevated cadmium levels in fish from Lake Powell, Arizona (station 93).

Columbia River System—Bottom sediments of the Willamette River and its numerous tributaries have cadmium concentrations ranging from 0.5 to 1 ppm (*73*). Concentrations of 2.5 ppm cadmium were found in sediment samples near the river mouth downstream from Portland, Oregon. Uniformity in sediment cadmium concentrations throughout the Williamette River basin suggests a geologic source of the metal. Active lead-zinc-silver mining in the Salmon River basin (station 43) may

account for the elevated concentrations in large-scale suckers (*23, 21*). High cadmium concentrations in fish from Grand Coulee, Washington (station 98), may reflect industrial waste from Spokane and activities of the Bunker Hill smelting complex at Kellogg, Idaho. Water from the South Fork of the Coeur d'Alene River has contained up to 0.45 ppm cadmium, corresponding to a cadmium transport of 240 lb cadmium/day (*22, 37*). The South Fork drains an area where thousands of tons of mine ground tailings of lead-zinc-silver ores were dumped decades ago (*15*). The Bunker Hill and associated smelters have tailing ponds extending for over 4 miles in the flood plain of the South Fork of the Coeur d'Alene River (*78*).

LEAD

Lead concentrations in freshwater fish (Table 4) ranged from 0.10 to 4.93 ppm and averaged 0.32 ppm. The trend in lead concentrations (Table 6) indicated no significant change from 1972 to 1976–77. The NPMP stations having concentration means at or above the 85th percentile of 0.44 ppm are discussed below.

Atlantic Coastal Streams—Segments of the Atlantic coastal streams where stations 2, 3, 4, and 53 are located contain many different types of industry. Industrial sources of lead could include those from metal finishing, brass manufacturing, lead alkyl manufacturing, primary and secondary lead smelting, coal combustion, and lead oxide manufacturing. River mud in the vicinity of a New Jersey zinc company, located on a tributary of the Delaware River, contained 0.13 percent lead (*78*). The St. Lawrence, New York, area contains a geologic source of lead; the Balmat mine, located there, ranks 18th in domestic output (*61*). The headwaters of the Hudson River may receive a lead flux from these ore deposits.

Great Lakes Drainage—Lake St. Clair (station 107), like the Atlantic coastal streams, is bordered by substantial industry and has a well-documented history of pollution (*13*). Lake St. Clair may receive urban lead aerosol fallout from the Detroit area, as well as effluents from numerous Detroit industries.

Mississippi River System—The Missouri-Oklahoma-Kansas vicinity is the location of stratabound deposits characteristically containing lead ores (*64*). Thus, in addition to point industrial sources of lead, the Verdigris River (station 78) and Red River (station 82) may receive lead from geologic origins. Fish residues at station 24 (Ohio River) may be affected by effluents (aerial fallout, tailing erosion, etc.) from a zinc company at Monaca, Pennsylvania. High concentrations at stations 88 and 89 (South Platte River), presumably reflect industrial discharges from Denver. The Pierre Shale of the Great Plains region contains low concentrations of lead and may provide a natural source of lead for stations 31, 86, 87, and 88 (*69*).

Columbia River System—Lead residues from fish at Grand Coulee, Washington (station 98), may be influenced by several lead sources affecting Franklin D. Roosevelt Lake. These are industrial effluents from Spokane, lead-zinc mining in Pend Oreille, Washington; natural stratabound deposits in Metaline Falls, Washington; and the mining-smelting complex at the South Fork of the Coeur d'Alene River. The Spokane water supply has had concentrations of copper and zinc above those allowed by Public Health Drinking Water Standards. The source is erosion and leaching of heavy metals from slag and tailing piles along the South Fork of the Coeur d'Alene River (*78*).

Hawaiian Streams—Waikele Stream (station 99) and its tributaries total 117 miles in length and drain a 45.7-square-mile watershed. Diversions for domestic and agricultural uses and highly permeable soils contribute to extreme variations in flows, which ranged from 0.02 to 13,600 ft^3/second over 24 years. Heavy urban runoff is characteristic of the flood flows in the lower reaches of the stream. Agricultural and residential use of lead arsenate has occurred in the drainage area, and vehicle sources of lead are prevalent. The Manoa-Palola stream system (station 100) drains an area of about 9.35 miles characterized by high vehicle density. Air quality data have reflected high levels of lead aerosols and other vehicle pollutants in the area. Lead arsenate has also been applied in the past for agricultural use and termite control (Lenhart, D. J. 1979. Regional Pesticide Specialist, Fish and Wildlife Service, U.S. Department of the Interior, Portland, Oregon, personal communication).

MERCURY

Mercury concentrations in fish in 1976–77 (Table 4) ranged from 0.01 to 0.84 ppm, and averaged 0.11 ppm. The decline of mercury levels in freshwater fish since 1972 (Table 6) is probably due to an overall reduction of industrial mercury emissions, coupled with a relative decrease in total domestic mercury consumption over the same period (*65*). NPMP monitoring stations having fish with mercury concentrations exceeding the 85th percentile (0.19 ppm; Table 7) are discussed below.

Atlantic Coastal Streams—The Penobscot and Kennebec River stations 1 and 51 have a long history of chloralkali and paper-pulping operations (Haines, T. A. 1979. Field Research Station Leader, Fish and Wildlife Service, Orono, Maine, personal communication). Unfiltered water samples from various sites in both rivers had total mercury levels in 1971 that equaled or exceeded the 0.2 μg/liter recommended by the National Academy of Sciences Water Quality Criteria (*35, 40*). U.S Geological Survey water quality data (retrieved through STORET)

also substantiated concentrations > 0.2 μg/liter; some values as high as 0.5 μg/liter were reported from the Penobscot River in 1978 (*74*). An old paper industry at Hartsville on a tributary to the Pee Dee River (station 56), most likely discharged mercury that became entrapped in sediments years ago. Georgia's Altamaha River (station 57) was studied intensively in 1970, and mercury residues of 1.0 ppm in largemouth bass were reported by the Georgia Water Control Board (*18*). A large pulp processing company is just above this sampling site (station 57), at Doctortown, Georgia.

Gulf Coast Streams—Alabama's Tombigbee River (station 14) and Alabama River (station 59) both exceeded the 85th percentile of 0.19 ppm, with fish from station 14 having a mean concentration of 0.33 ppm. There were two chloralkali plants contaminating the Tombigbee in the early 1970's (*12*)—one at McIntosh and the other at river mile 26 (Smith, B. W. 1970. Assistant Chief of Fisheries Section, Alabama Game and Fish Commission, personal communication). Mercury concentrations have been relatively high in fish from the Alabama River throughout the NPMP samplings (*20, 87*), but the source of mercury has not been identified.

Great Lakes Drainage—The Lake St. Clair (station 107) mercury problem originated from chloralkali operations at Sarnia, Ontario, and is well documented (*12, 13*). In 1970, mercury concentrations in fillets from four species of Lake St. Clair fish exceeded 1 ppm (*67*). Another Great Lakes station at which fish had mercury concentrations greater than 0.19 ppm was station 22 on Lake Superior. The Ontario Department of Lands and Forests reported mercury concentrations > 0.5 ppm in the muscle of fish from different parts of Lake Superior in 1970 (*12*). The source was presumably chloralkali plants at Marathon and Thunder Bay. The St. Lawrence River (station 66) was listed by FDA as seriously contaminated in 1970. Mercury cell chloralkali plants are still operating along the river at Cornwall, New York, and Beauharnois and Shawinigan, Quebec (*13*).

Mississippi River System—In the Red River (station 34), which drains into Lake Winnipeg in Manitoba Province, the 0.28-ppm mercury concentration in fish exceeds the 85th percentile. The source of mercury has not been identified. Station 70 at Metropolis, Illinois, is directly downstream from the confluence of the Kentucky and Ohio Rivers. Two mercury cell chloralkali plants are located at Calvert City, Kentucky, near the mouth of the Kentucky River (*13*).

Interior Basins—Pre-1900 gold and silver milling operations of the Nevada Comstock Lode introduced substantial amounts of mercury into the Truckee and Carson River drainage systems; in bottom sediments, total mercury concentrations were as high as 20 ppm in 1971 (*48*). Concentrations in fish (0.50–2.72 ppm) were highest in white bass, a piscivorous species from Lahontan Reservoir. Bottom sediments in the Truckee River basin (station 37) contained greater than background mercury concentrations, as a result of ore milling activity in the Washoe Valley (*72*).

Columbia River System—The Columbia River system, including the tributary Yakima, Willamette, Snake, and Salmon Rivers is on the East Pacific Rise, the location of major mercury deposits (cinnabar) in the Western Hemisphere. There are secondary mercury mining operations in Washington and Idaho, as well as gold mining, where mercury was used to recover gold from its ores by amalgamation (*12*). In 77 percent of northern squawfish samples from the Salmon River, the axial musculature contained mercury concentrations higher than 0.5 ppm (*8*).

ARSENIC

Arsenic concentrations in fish (Table 4) ranged from 0.05 to 2.92 ppm and averaged 0.27 ppm. The increase in arsenic in freshwater fish since 1972 (Table 6) may be the result of dissemination by air pollution, smelter solid waste disposal, and continued use of arsenical pesticides. Contamination sources for arsenic are suggested here for the $\geqslant$ 85th percentile ($\geqslant 0.38$ ppm) NPMP stations.

Atlantic Coastal Streams—No specific industrial source for arsenic in the Hudson River (station 3) is known. Extensive industrialization of the river, however, presumably accounts for high arsenic concentrations in fish. In Georgia and South Carolina, substantial numbers of cotton farms (*71*) border the Savannah River and its tributaries (station 10). Arsenicals were used extensively on cotton from the early 1940's through the middle 1960's in Georgia (Winstead, E. E. 1979. Assistant Commissioner, Georgia Department of Agriculture, Atlanta, personal communication). Agricultural runoff of applied arsenicals would provide a persistent source of arsenic to sediment beds.

Gulf Coast Streams—In 1976, the Southern Plains states had more than 5 million acres planted in cotton, compared with slightly over 3 million for the Delta states (*57*). The highest concentration of cotton farms in the Southern Plains occurs along the Rio Grande Valley and western Texas Panhandle. Stations 82 and 60 are located on the Red and Brazos River systems, respectively, that drain these cotton-growing areas. Arsenic acid was the most heavily applied arsenical in 1976 in the Southern Plains, followed by sodium cacodylate and DSMA (*57*).

Great Lakes Drainage—Relatively high arsenic levels have been reported in many areas of Lake Michigan. In the southern portion (stations 21 and 105), sediment

concentrations have reached values as high as 30 ppm (*50*). In the northern portion (station 104), arsenic has accumulated in ferromanganese nodules that exist in the Green Bay area (*50*). One of the more striking cases of arsenic pollution involves the Ansul Co. of Marinette, Wisconsin (located close to the Menominee River, which empties into Green Bay). That company was a major manufacturer of methanearsonic acid (MAA) and cacodylic acid (CA), both arsenical herbicides (*2*). The company stored arsenic-contaminated sodium chloride and sodium sulfate manufacturing by-products in unprotected salt piles on the bank of the river. Precipitation runoff from the piles produced levels of $>$ 200 ppm arsenic in river sediments. Groundwater below the piles had total arsenic concentrations in excess of 6,000 ppm. Sediment levels adjacent to the salt piles were 2 percent arsenic by weight. As a result, the Menominee River is responsible for contributing 30–50 tons of arsenic per year to Lake Michigan (*2*). Marsh (*36*) concluded that there was a definite accumulation of lead and arsenic in and around Grand Traverse Bay in northeast Lake Michigan. Lead arsenate pesticides, used as orchard spray, accounted for all of the arsenic and about half of the lead buildup.

In contrast to Lake Michigan, specific sources of arsenic in Lake Superior (stations 22, 102, and 103) and the St. Lawrence River (station 66) were not readily apparent. A nonpoint source affecting all of the Great Lakes was mentioned by Traversy et al. (*24*), who reported that arsenic levels in precipitation were higher than those in water from the Great Lakes and surrounding rivers. The elevated arsenic precipitation levels were especially prevalent at or near highly industrialized locations such as Toronto, Sarnia, and Hamilton.

A mine and copper smelting facility at White Pine, Michigan, is on the Mineral River, which drains into southern Lake Superior. Wash water and smelter runoff flow into Mineral River, as well as uncontrolled erosion from slag piles and tailing pond outfalls (*78*). Any of these effluents could contain substantial amounts of arsenic.

Arsenic concentrations in whole fish from the Great Lakes have been reported by Lucas et al. (*33*) and Traversy et al. (*24*) for the period from 1968 to 1971. Lucas et al., who analyzed 19 fish of 3 species, found a mean of 0.16 ppm arsenic, whereas Traversy et al., who analyzed 43 whole fish samples of 15 species, reported a mean of 0.063 ppm. The present authors report a mean arsenic concentration level of 0.72 ppm for fish from Great Lakes stations, including data from 33 sample composites of 10 different species. Although species, location, and methodology differences cannot be ruled out, the mean differences suggest a significant increase in arsenic concentrations in Great Lakes fish from 1970 to 1977. The arsenic levels in the present report are similar to those found by the Upper Lakes Reference Group (*84*). At stations where arsenic seems to be higher than background levels—i.e., greater than the 85th percentile (0.38 ppm)—concentrations in bloaters (*Coregonus* spp.) tended to be about double those in lake trout (stations 21, 102, 104, and 105). The tendency of bloaters to concentrate arsenic is apparent from other work (*11, 84*) and may be related to a primary diet of zooplankton, which has been shown to bioconcentrate arsenic (*80*).

Columbia River System—Stations 42–46 had mean arsenic residues above the calculated 85th percentile (Table 7). Such a preponderance of "high" stations in a relatively small geographical area gives credence to the following four possible sources of arsenic pollution:

1. Volcanic eruptions in the central Cascade area during the Eocene epoch resulted in a large accumulation of volcanic deposits referred to as the Fisher formation. In some instances, arsenic and perhaps boron were a part of the pyroclastic debris that formed the so-called Fisher rocks. Arsenic released from Fisher rocks by percolating subsurface water has resulted in arsenic groundwater contamination in some areas of the southern Willamette Valley (*31*). This geologic source of arsenic may be present in other areas of the Columbia River system.

2. A copper-smelting facility at Tacoma, Washington, and the lead-smelting refineries located at Kellogg, Idaho (Bunker Hill), and East Helena, Montana, may provide airborne sources of arsenic to the Columbia River system. Soil and water pollution are possible through smelter solid waste disposal (*78*).

3. Active mining of copper, lead, and gold may present mine water and mine tailing disposal problems, because arsenic is found in association with these base-metal ores.

4. In the headwater regions of the Yakima, monosodium methanearsonate (MSMA) and cacodylic acid are currently used for thinning in forestry (Gregory, S. A. 1979. Field Station Leader, Columbia National Fisheries Research Laboratory, Fish and Wildlife Service, Corvallis, Oregon, personal communication).

SELENIUM

Concentrations of selenium in fish ranged from 0.05 to 2.87 ppm and averaged 0.56 ppm. Stations having mean concentration levels exceeding the 85th percentile (0.82 ppm) are discussed below.

Atlantic Coastal Streams—Fish from station 4 on the Delaware River at Camden, New Jersey, had concentrations exceeding the 85th percentile, not only for

selenium (0.82 ppm), but also for cadmium and lead. The elevated trace element concentrations in these fish probably reflect the highly industrialized character of the Delaware River.

Mississippi River System—The Big Horn (station 84) and Yellowstone (station 85) River tributaries of the Missouri River are closely associated with Montana's Fort Union coal formation and outcroppings of phosphate beds in Montana and Wyoming (*45, 71*). Selenium concentrations in fish from these rivers may result from a geologic source of the element. Selenium sources to the aquatic environment at the South Platte River near Denver (station 88) may be industrial effluents or deposits of coal, barite, and sulfur ore (*71*). Phosphate bed outcroppings are located along the Kansas-Missouri border close to station 90 on the Kansas River (*71*). Selenium concentrations in fish from stations 80 and 31 may also reflect a geologic source of the element, primarily sedimentary rocks associated with the Pierre formation.

California Streams—The source of selenium in fish at station 40 on the San Joaquin River is unknown. Selocide, a selenium-containing pesticide, was registered for use on citrus in California in the 1960's (*41*). This material may have been applied in the San Joaquin River valley and, if so, could still be a source of selenium to the aquatic environment.

Summary

Primary sources of the trace elements to the aquatic environment follow:

Cadmium: electroplating industry, zinc-lead-copper smelting and refining, phosphate fertilizers, sulfide ore-mining activities.

Lead: combustion of gasoline, lead-zinc-copper smelting operations, sulfide ore-mining activities, coal combustion.

Mercury: pre-1975 chloralkali industry, pre-1972 paper-pulping operations, synthetic fiber industries, coal combustion.

Arsenic: copper-lead-gold smelting and refining, coal combustion, smelter solid waste disposal, arsenical pesticides, geologic.

Selenium: geologic, industrial.

Once in the environment, there is evidence that all five trace elements may undergo biologically mediated transformation reactions that yield organometallic compounds that are routed through the food chain.

Trace element concentrations (mg/kg wet weight) in whole fish in 1976–77 follow:

Metal	Range	Geometric Mean	Median	85th Percentile
Cadmium	0.01–1.04	0.07	0.05	0.11
Lead	0.10–4.92	0.32	0.19	0.44
Mercury	0.01–0.84	0.11	0.09	0.19
Arsenic	0.05–2.92	0.27	0.25	0.38
Selenium	0.05–2.87	0.56	0.50	0.82

Temporal trends in whole-fish trace element concentrations (mg/kg wet weight) from 1972 to 1976–77 were as follows: cadmium, significant decline; lead, no significant difference; mercury, significant decline; arsenic, significant increase; and selenium, no significant difference (1972 vs. 1977).

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FOOD AND FEED

Pesticide, Heavy Metal, and Other Chemical Residues in Infant and Toddler Total Diet Samples—(II)—August 1975–July 1976 [1]

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ABSTRACT

The Food and Drug Administration, U.S. Department of Health and Human Services, initiated the Total Diet Study in 1964 to monitor residues of pesticides and other chemicals ingested in the average diet of the United States' heartiest eater, the young adult male. In August 1974, one-third of the adult market baskets were replaced with infant and toddler market baskets. Averages and ranges of residues for this second in a series of infant and toddler baskets, for August 1975–July 1976, are reported. Included are results of determinations for zinc, cadmium, lead, selenium, arsenic, and mercury. Results of recovery studies conducted with compounds of each residue type are also presented.

Introduction

The Food and Drug Administration (FDA), U.S. Department of Health and Human Services (formerly U.S. Department of Health, Education, and Welfare), has been monitoring the United States' total diet since 1964 (*1–6, 11–17*). The program began with surveillance of crops for fission products from atmospheric tests of thermonuclear weapons. Later, the emphasis was directed toward pesticide residues in foods. For several years the program focused on the total diet of the 16- to 19-year-old male, statistically the United States' heartiest eater. In August 1974, 10 of the 30 market baskets were replaced with the total diet of 6-month-old infants and 2-year-old toddlers. The 10 market baskets were collected in 10 cities that ranged in population from fewer than 50,000 to one million or more.

Foods in each of 11 broad classes, as listed in Table 1, were prepared separately, composited into a slurry, and analyzed for organochlorine and organophosphorus pesticides, carbaryl, herbicides, metals, and a few industrial chemicals. Methodologies included atomic absorption spectroscopy, fluorometry, gas chromatography, thin-layer chromatography, mass spectroscopy, and established extraction and cleanup techniques (*7, 8, 10, 18, 19*). Except for the water composite (*9*), quantitation limits and instrumental conditions were the same as those described for the adult market baskets.

TABLE 1. *Commodity classes of infant and toddler foods analyzed for pesticides, metals, and other chemical residues, August 1975–July 1976*

Key	Food Class
I	Drinking water [1]
II	Whole milk, fresh [1]
III	Other dairy and substitutions, infant
	Other dairy and substitutions, toddler
IV	Meat, fish, and poultry, infant
	Meat, fish, and poultry, toddler
V	Grain and cereal products, infant
	Grain and cereal products, toddler
VI	Potatoes [1, 2]
VII	Vegetables, infant
	Vegetables, toddler
VIII	Fruit and fruit juice, infant
	Fruit and fruit juice, toddler
IX	Oils and fats [1, 3]
X	Sugar and adjuncts, infant
	Sugar and adjuncts, toddler
XI	Beverages [1, 4]

NOTE: Use key with Table 3.
[1] Because of similarity between infant and toddler diets, single determinations for certain classes of food are made and reported for both.
[2] No infant composite for western region.
[3] No infant composite from the north-central, western, and southern regions.
[4] No infant composite from north-central region.

Results

The infant composites of the present series contained 301 residues of 31 compounds, with 51 residues at the trace level. In comparison, the first infant composites reported last year contained 306 residues of 28 compounds; 121 were reported at the trace level.

Toddler composites of the present series showed 473 residues of 38 compounds; 76 were present in trace amounts. The first toddler series last year reported 468 residues of 30 compounds with 179 at the trace level.

[1] Food and Drug Administration, Kansas City District Office Laboratory, 1009 Cherry St., Kansas City, MO 64106

The chemical compounds found, the number of findings, and the range for each are listed in Table 2. The frequency of occurrence of each compound by food class is presented in Table 3. Table 4 shows the level of every residue found within each food class. The averages given in Table 4 are based on the total number of composites examined for that food class. Trace values were treated as zero in calculating these averages. Table 5 shows the intake of pesticide and industrial chemical residues in terms of μg/kg body weight/day, and Table 6 shows the intake of six metals in terms of μg/day (mg/day for zinc). For comparison, the findings for the 1975 fiscal year are also shown. The most common residues and their average levels are discussed below for each of the 11 food classes. No findings have been corrected for recoveries.

DRINKING WATER

Infant and Toddler—Tap water samples analyzed were taken from the same location as the market basket. The result of a single analysis is that reported for both infant and toddler composites. The water was used to prepare other market basket items requiring dilution or addition of water. Only two metal residues were found: four samples contained zinc averaging 0.170 ppm for the series, and one sample contained cadmium, averaging 0.002 ppm.

WHOLE MILK, FRESH

Infant and Toddler—This composite was common to both diets. Averages for chlorinated pesticide residues included 0.001 ppm *p,p'*-DDE; trace α-BHC (hexachlorocyclohexane); trace dieldrin; and a trace of heptachlor epoxide. Averages for the three metals found were 6.80 ppm zinc; 0.007 ppm cadmium; and 0.002 ppm selenium. A trace of PCP was detected in one composite.

OTHER DAIRY AND SUBSTITUTIONS

Infant—The variation in infants' and toddlers' diets becomes apparent with these composites. Dieldrin was found in four composites averaging trace for the series. Averages of other organochlorines were traces of α-BHC, heptachlor epoxide, methoxychlor, and *p,p'*-DDE. A trace of PCP was found in one composite. Averages for metal residues were 5.38 ppm zinc, 0.004 ppm cadmium, and 0.013 ppm lead.

Toddler—All 10 composites contained dieldrin, averaging 0.003 ppm, and α-BHC, averaging 0.002 ppm. Averages of other pesticide residues included 0.004 ppm *p,p'*-DDE, 0.001 ppm heptachlor epoxide, and traces of octachlor epoxide, methoxychlor, HCB, and lindane. A trace of PCP was found in one composite. Averages of metal residues included 5.94 ppm zinc, 0.016 ppm selenium, 0.013 ppm cadmium, 0.006 ppm lead, and a trace of mercury.

TABLE 2. *Chemical and metal residues found in infant and toddler food composites from 10 United States cities—August 1975–July 1976*

Chemical Found	No. of Composites With Residues	No. of Positive Composites With Residues Reported as Trace [1]	Range, ppm
INFANT			
Zinc	85	0	0.100–36.3
Cadmium	59	0	0.005–0.130
Lead	35	0	0.050–0.290
Selenium	21	0	0.020–0.250
p,p'-DDE [2]	17	8	0.001–0.019
Dieldrin	15	7	0.001–0.003
Malathion	12	0	0.003–0.049
α-BHC	9	7	0.0006–0.001
Heptachlor epoxide	6	6	T
Mercury	4	0	0.002–0.016
PCP	4	4	T
CIPC	3	0	0.023–0.469
Arsenic	3	0	0.040–0.090
Dichloran	3	0	0.006–0.007
Endosulfan [2]	3	2	0.037
HCB	3	3	T
Endrin	2	1	0.011
Lindane	2	0	0.004
Toxaphene	2	2	T
Octachlor epoxide	2	2	T
TCNB	1	0	0.026
p,p'-DDT [2]	1	1	T
Chlordane	1	1	T
Diazinon	1	0	0.010
Methoxychlor	1	1	T
Parathion	1	1	T
Ethion	1	1	T
Fonofos	1	1	T
Carbaryl	1	1	T
TCTA	1	1	T
Perthane	1	1	T
TODDLER			
Zinc	100	0	0.100–34.0
Cadmium	77	0	0.005–0.120
Lead	35	0	0.050–0.350
Dieldrin	31	6	0.001–0.007
α-BHC	29	12	0.0006–0.003
Selenium	27	0	0.020–0.300
p,p'-DDE [2]	26	4	0.001–0.022
Heptachlor epoxide	20	13	0.001–0.007
Malathion	16	0	0.005–0.187
Lindane	15	3	0.001–0.007
HCB	14	8	0.0007–0.016
Arsenic	11	0	0.030–0.290
Octachlor epoxide	10	9	0.004
Mercury	7	0	0.002–0.020
PCP	6	5	0.070
CIPC	5	0	0.023–0.469
PCA	4	0	0.002–0.044
Pentachlorobenzene	4	1	0.001–0.009
Toxaphene	4	3	0.075
Dichloran	3	0	0.012–0.026
PCNB	3	0	0.001–0.005
Diazinon	3	1	0.001–0.003
Chlordane	3	2	0.137
Parathion	3	3	T
p,p'-DDT [2]	2	1	0.005
p,p'-TDE [2]	2	1	0.002
Fonofos	2	0	0.001–0.002
Ronnel	1	0	0.002
Endrin	1	0	0.009
2,4-D	1	0	0.025
TCNB	1	0	0.026
trans-Nonachlor	1	0	0.006
PCTA	1	0	0.003
TCTA	1	1	T
Ethion	1	1	T
o-Phenylphenol	1	1	T
Methoxychlor	1	1	T
PCP methyl ether	1	0	0.002

[1] Chemicals detected by the specific analytical methodology below the limit of quantitation were confirmed qualitatively and reported as trace (T). The limits of quantitation vary with residues and food classes.

[2] Other isomers also included in reporting.

[3] Reportings include isomers I and II and sulfate.

TABLE 3. *Frequency of occurrence of chemical and metal residues, by food class, in infant and toddler food composites from 10 United States cities—August 1975–July 1976*

Chemical	Food Class [1]										
	I	II	III	IV	V	VI	VII	VIII	IX	X	XI
NUMBER OF OCCURRENCES IN INFANT FOODS											
Zinc	4	10	10	10	10	8	10	9	2	7	5
Cadmium	1	4	4	7	10	8	10	6	2	6	1
Lead	0	0	1	8	6	6	8	5	0	0	1
Selenium	0	1	0	10	10	0	0	0	0	0	0
p,p'-DDE	0	3	4	5	0	3	2	0	0	0	0
Dieldrin	0	2	4	6	0	2	0	0	1	0	0
Malathion	0	0	0	0	10	0	0	0	2	0	0
α-BHC	0	3	5	1	0	0	0	0	0	0	0
Heptachlor epoxide	0	1	3	0	0	0	0	0	2	0	0
Mercury	0	0	0	2	0	0	1	0	0	1	0
PCP	0	1	1	0	1	0	0	1	0	0	0
CIPC	0	0	0	0	0	3	0	0	0	0	0
Arsenic	0	0	0	0	3	0	0	0	0	0	0
Dichloran	0	0	0	0	0	1	0	2	0	0	0
Endosulfan	0	0	0	0	0	1	1	1	0	0	0
HCB	0	0	0	2	0	1	0	0	0	0	0
Endrin	0	0	0	0	0	0	0	0	2	0	0
Lindane	0	0	0	0	2	0	0	0	0	0	0
Toxaphene	0	0	0	0	0	0	0	0	2	0	0
Octachlor epoxide	0	0	0	2	0	0	0	0	0	0	0
TCNB	0	0	0	0	0	1	0	0	0	0	0
p,p'-DDT	0	0	0	0	0	0	1	0	0	0	0
Chlordane	0	0	0	0	0	0	0	0	1	0	0
Diazinon	0	0	0	0	1	0	0	0	0	0	0
Methoxychlor	0	0	1	0	0	0	0	0	0	0	0
Parathion	0	0	0	0	0	0	1	0	0	0	0
Ethion	0	0	0	0	0	0	0	1	0	0	0
Fonofos	0	0	0	0	0	0	0	0	1	0	0
Carbaryl	0	0	0	0	0	0	0	1	0	0	0
TCTA	0	0	0	0	0	1	0	0	0	0	0
Perthane	0	0	0	0	0	0	0	1	0	0	0
NUMBER OF OCCURRENCES IN TODDLER FOODS											
Zinc	4	10	10	10	10	10	10	9	10	10	7
Cadmium	1	4	7	9	10	10	10	4	10	10	2
Lead	0	0	1	3	5	6	6	5	6	2	1
Dieldrin	0	2	10	10	0	2	2	0	5	0	0
α-BHC	0	3	10	8	0	0	2	0	0	6	0
Selenium	0	1	4	9	10	0	0	0	2	1	0
p,p'-DDE	0	3	8	10	0	3	1	0	1	0	0
Heptachlor epoxide	0	1	8	9	0	0	0	0	2	0	0
Malathion	0	0	0	0	10	0	0	0	6	0	0
Lindane	0	0	1	4	0	1	3	0	0	6	0
HCB	0	0	5	4	0	1	0	0	4	0	0
Arsenic	0	0	0	7	2	0	0	0	1	1	0
Octachlor epoxide	0	0	6	4	0	0	0	0	0	0	0
Mercury	0	0	1	6	0	0	0	0	0	0	0
PCP	0	1	1	0	1	0	1	1	0	1	0
CIPC	0	0	0	0	0	5	0	0	0	0	0
PCA	0	0	0	0	0	0	0	0	4	0	0
Pentachlorobenzene	0	0	0	0	0	0	0	0	4	0	0
Toxaphene	0	0	0	0	0	0	0	0	4	0	0
Dichloran	0	0	0	0	0	0	0	3	0	0	0
PCNB	0	0	0	0	0	0	0	0	3	0	0
Diazinon	0	0	0	0	3	0	0	0	0	0	0
Chlordane	0	0	0	0	0	0	0	0	3	0	0
Parathion	0	0	0	0	0	0	1	0	2	0	0
p,p'-DDT	0	0	0	2	0	0	0	0	0	0	0
p,p'-TDE	0	0	0	2	0	0	0	0	0	0	0
Fonofos	0	0	0	0	0	0	0	0	2	0	0
Ronnel	0	0	0	1	0	0	0	0	0	0	0
Endrin	0	0	0	0	0	0	0	0	1	0	0
2,4-D	0	0	0	0	0	0	0	0	0	1	0
TCNB	0	0	0	0	0	1	0	0	0	0	0
trans-Nonachlor	0	0	0	1	0	0	0	0	0	0	0
PCTA	0	0	0	0	0	0	0	0	1	0	0
TCTA	0	0	0	0	0	1	0	0	0	0	0
Ethion	0	0	0	0	0	0	0	1	0	0	0
o-Phenylphenol	0	0	0	0	0	0	0	1	0	0	0
Methoxychlor	0	0	1	0	0	0	0	0	0	0	0
PCP methyl ether	0	0	0	0	0	0	0	0	1	0	0

[1] See Table 1 for key to food classes.

MEAT, FISH, AND POULTRY

Infant—These infant composites contained among the highest number of residues. Metals dominated 37 of the 53 residues, with averages of 16.8 ppm zinc, 0.094 ppm selenium, 0.087 ppm lead, 0.023 ppm cadmium, and 0.002 ppm mercury. Of the 16 nonmetals, dieldrin was found in six composites averaging a trace for the series; *p,p'*-DDE, averaging 0.003 ppm for the series, was found in five composites; HCB, octachlor epoxide, and α-BHC averaged a trace.

Toddler—Ninety-nine residues were found. All 10 composites contained residues of *p,p'*-DDE, ranging from 0.002 to 0.016 ppm, and dieldrin, ranging from 0.001 to 0.003 ppm. Heptachlor epoxide, *p,p'*-TDE, *p,p'*-DDT, hexachlorobenzene (HCB), α-BHC, lindane, octachlor epoxide, ronnel, and *trans*-nonachlor averaged a trace for the series. Average metal residues included 27.5 ppm zinc and 0.180 ppm selenium. Lesser amounts of arsenic, lead, cadmium, and mercury, averaging 0.092, 0.030, 0.010, and 0.006 ppm, respectively, were also found.

GRAIN AND CEREAL PRODUCTS

Infant—All 10 composites contained malathion, ranging from 0.003 to 0.049 ppm and averaging 0.020 ppm. There was one report of diazinon at the 0.010 ppm level, and only traces of lindane and PCP were found. Averages for the metals were 14.6 ppm zinc, 0.189 ppm selenium, 0.046 ppm lead, 0.027 ppm cadmium, and 0.018 ppm arsenic.

Toddler—Like the infant composites, these toddler composites all contained malathion residues, ranging from 0.006 to 0.025 ppm and averaging 0.012 ppm. Diazinon, averaging trace amounts, was found in three composites. Averages for the metals found were 10.3 ppm zinc, 0.179 ppm selenium, 0.065 ppm lead, 0.026 ppm cadmium, and 0.008 ppm arsenic.

POTATOES

Infant—The chlorinated herbicide CIPC was found in three composites, ranging from 0.023 to 0.469 ppm and averaging 0.073 ppm for the series. TCNB averaged 0.003 ppm for the series. Other organochlorine pesticides averaging trace amounts included *p,p'*-DDE, dieldrin, dichloran, endosulfan, TCTA, and HCB. Of the metals found, zinc averaged 2.72 ppm, lead averaged 0.061 ppm, and cadmium averaged 0.048 ppm.

Toddler—The chlorinated herbicide CIPC was found in five composites, ranging from 0.023 to 0.469 ppm and averaging 0.156 ppm for the series. TCNB was found in one composite at 0.026 ppm, and dieldrin, lindane, *p,p'*-DDE, TCTA, and HCB averaged a trace. The metals zinc and cadmium averaged 3.92

TABLE 4. *Levels of chemical and metal residues, by food class, in infant and toddler food composites from 10 United States cities—August 1975–July 1976*

Chemical	Residue, ppm	
	Infant	Toddler
I. WATER		
ZINC		
Average	0.170	0.170
Positive composites		
Total number	4	4
Number reported as trace	0	0
Range	0.200–0.600	0.200–0.600
CADMIUM		
Average	0.002	0.002
Positive composites		
Total number	1	1
Number reported as trace	0	0
Range	0.02	0.020
II. WHOLE MILK, FRESH		
ZINC		
Average	6.80	6.80
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	3.30–28.8	3.30–28.8
α-BHC		
Average	T	T
Positive composites		
Total number	3	3
Number reported as trace	2	2
Range	0.0006	0.0006
p,p'-DDE		
Average	0.001	0.001
Positive composites		
Total number	3	3
Number reported as trace	1	1
Range	0.002–0.005	0.002–0.005
DIELDRIN		
Average	T	T
Positive composites		
Total number	2	2
Number reported as trace	0	0
Range	0.001	0.001
CADMIUM		
Average	0.007	0.007
Positive composites		
Total number	4	4
Number reported as trace	0	0
Range	0.005–0.040	0.005–0.040
PCP		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
HEPTACHLOR EPOXIDE		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
SELENIUM		
Average	0.002	0.002
Positive composites		
Total number	1	1
Number reported as trace	0	0
Range	0.02	0.020
III. OTHER DAIRY AND SUBSTITUTIONS		
ZINC		
Average	5.38	5.94
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	2.90–8.50	1.60–14.6
HEPTACHLOR EPOXIDE		
Average	T	0.001
Positive composites		
Total number	3	8
Number reported as trace	3	3
Range	T	0.001–0.003
DIELDRIN		
Average	T	0.003
Positive composites		
Total number	4	10
Number reported as trace	2	0
Range	0.001	0.001–0.005
METHOXYCHLOR		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
CADMIUM		
Average	0.004	0.013
Positive composites		
Total number	4	7
Number reported as trace	0	0
Range	0.005–0.020	0.006–0.070
α-BHC		
Average	T	0.002
Positive composites		
Total number	5	10
Number reported as trace	4	0
Range	0.001	0.001–0.003
p,p'-DDE		
Average	T	0.004
Positive composites		
Total number	4	8
Number reported as trace	4	1
Range	T	0.001–0.022
LEAD		
Average	0.013	0.006
Positive composites		
Total number	1	1
Number reported as trace	0	0
Range	0.130	0.060
PCP		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
SELENIUM		
Average		0.016
Positive composites		
Total		4
Number reported as trace		0
Range		0.030–0.050
OCTACHLOR EPOXIDE		
Average		T
Positive composites		
Total number		6
Number reported as trace		6
Range		T
MERCURY		
Average		T
Positive composites		
Total number		1
Number reported as trace		0
Range		0.002
HCB		
Average		T
Positive composites		
Total number		5
Number reported as trace		4
Range		0.0007
LINDANE		
Average		T
Positive composites		
Total number		1
Number reported as trace		0
Range		0.002
IV. MEAT, FISH, AND POULTRY		
ZINC		
Average	16.84	27.52
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	8.90–36.3	11.3–34.0

TABLE 4 (cont'd.). *Levels of chemical and metal residues, by food class, in infant and toddler food composites from 10 United States cities—August 1975–July 1976*

Chemical	Residue, ppm	
	Infant	Toddler
SELENIUM		
Average	0.094	0.180
Positive composites		
Total number	10	9
Number reported as trace	0	0
Range	0.040–0.220	0.110–0.300
LEAD		
Average	0.087	0.030
Positive composites		
Total number	8	3
Number reported as trace	0	0
Range	0.050–0.180	0.050–0.190
CADMIUM		
Average	0.023	0.010
Positive composites		
Total number	7	9
Number reported as trace	0	0
Range	0.010–0.100	0.008–0.020
MERCURY		
Average	0.002	0.006
Positive composites		
Total number	2	6
Number reported as trace	0	0
Range	0.004–0.016	0.005–0.020
DIELDRIN		
Average	T	0.002
Positive composites		
Total number	6	10
Number reported as trace	4	1
Range	0.002–0.003	0.001–0.003
HCB		
Average	T	T
Positive composites		
Total number	2	4
Number reported as trace	2	2
Range	T	0.0007–0.0009
α-BHC		
Average	T	T
Positive composites		
Total number	1	8
Number reported as trace	1	7
Range	T	0.0007
p,p'-DDE		
Average	0.003	0.005
Positive composites		
Total number	5	10
Number reported as trace	2	0
Range	0.002–0.019	0.002–0.016
OCTACHLOR EPOXIDE		
Average	T	T
Positive composites		
Total number	2	4
Number reported as trace	2	3
Range	T	0.004
ARSENIC		
Average		0.092
Positive composites		
Total number		7
Number reported as trace		0
Range		0.060–0.290
HEPTACHLOR EPOXIDE		
Average		T
Positive composites		
Total number		9
Number reported as trace		8
Range		0.002
p,p'-TDE		
Average		T
Positive composites		
Total number		2
Number reported as trace		1
Range		0.002
p,p'-DDT		
Average		T
Positive composites		
Total number		2
Number reported as trace		1
Range		0.005
LINDANE		
Average		T
Positive composites		
Total number		4
Number reported as trace		3
Range		0.003
RONNEL		
Average		T
Positive composites		
Total number		1
Number reported as trace		0
Range		0.002
trans-NONACHLOR		
Average		T
Positive composites		
Total number		1
Number reported as trace		0
Range		0.006

V. GRAIN AND CEREAL PRODUCTS		
ZINC		
Average	14.6	10.29
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	6.8–20.3	6.30–18.9
SELENIUM		
Average	0.189	0.179
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	0.140–0.250	0.080–0.290
CADMIUM		
Average	0.027	0.026
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	0.020–0.050	0.018–0.040
ARSENIC		
Average	0.018	0.008
Positive composites		
Total number	3	2
Number reported as trace	0	0
Range	0.040–0.090	0.030–0.050
MALATHION		
Average	0.020	0.012
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	0.003–0.049	0.006–0.025
LEAD		
Average	0.046	0.065
Positive composites		
Total number	6	5
Number reported as trace	0	0
Range	0.050–0.100	0.060–0.210
PCP		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
LINDANE		
Average	T	
Positive composites		
Total number	2	
Number reported as trace	0	
Range	0.004	
DIAZINON		
Average	0.001	T
Positive composites		
Total number	1	3
Number reported as trace	0	1
Range	0.010	0.001–0.003

TABLE 4 (cont'd.). *Levels of chemical and metal residues, by food class, in infant and toddler food composites from 10 United States cities—August 1975–July 1976*

Chemical	Residue, ppm	
	Infant	Toddler
VI. POTATOES		
ZINC		
Average	2.72	3.92
Positive composites		
Total number	8	10
Number reported as trace	0	0
Range	0.30–4.30	0.300–8.20
LEAD		
Average	0.061	0.078
Positive composites		
Total number	6	6
Number reported as trace	0	0
Range	0.050–0.120	0.050–0.350
CADMIUM		
Average	0.048	0.045
Positive composites		
Total number	8	10
Number reported as trace	0	0
Range	0.020–0.130	0.010–0.120
DIELDRIN		
Average	T	T
Positive composites		
Total number	2	2
Number reported as trace	1	1
Range	0.002	0.002
LINDANE		
Average		T
Positive composites		
Total number		1
Number reported as trace		0
Range		0.007
p,p'-DDE		
Average	T	T
Positive composites		
Total number	3	3
Number reported as trace	1	1
Range	0.001–0.002	0.002
DICHLORAN		
Average	T	
Positive composites		
Total number	1	
Number reported as trace	0	
Range	0.006	
ENDOSULFAN		
Average	T	
Positive composites		
Total number	1	
Number reported as trace	1	
Range	T	
CIPC		
Average	0.073	0.156
Positive composites		
Total number	3	5
Number reported as trace	0	0
Range	0.023–0.469	0.023–0.469
TCNB		
Average	0.003	0.003
Positive composites		
Total number	1	1
Number reported as trace	0	0
Range	0.026	0.026
TCTA		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
HCB		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T

Chemical	Residue, ppm	
	Infant	Toddler
VII. VEGETABLES		
ZINC		
Average	3.66	3.49
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	1.80–6.40	0.600–4.70
LEAD		
Average	0.078	0.081
Positive composites		
Total number	8	6
Number reported as trace	0	0
Range	0.060–0.240	0.070–0.200
CADMIUM		
Average	0.043	0.026
Positive composites		
Total number	10	10
Number reported as trace	0	0
Range	0.010–0.120	0.017–0.060
MERCURY		
Average	T	
Positive composites		
Total number	1	
Number reported as trace	0	
Range	0.002	
PARATHION		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
p,p'-DDE		
Average	T	T
Positive composites		
Total number	2	1
Number reported as trace	0	0
Range	0.002–0.004	0.002
p,p'-DDT		
Average	T	
Positive composites		
Total number	1	
Number reported as trace	1	
Range	T	
ENDOSULFAN		
Average	0.004	
Positive composites		
Total number	1	
Number reported as trace	0	
Range	0.037	
PCP		
Average		T
Positive composites		
Total number		1
Number reported as trace		1
Range		T
LINDANE		
Average		T
Positive composites		
Total number		3
Number reported as trace		0
Range		0.001–0.002
DIELDRIN		
Average		T
Positive composites		
Total number		2
Number reported as trace		1
Range		0.005
α-BHC		
Average		T
Positive composites		
Total number		2
Number reported as trace		0
Range		0.002

TABLE 4 (cont'd.). *Levels of chemical and metal residues, by food class, in infant and toddler food composites from 10 United States cities—August 1975–July 1976*

Chemical	Residue, ppm	
	Infant	Toddler
VIII. FRUITS AND FRUIT JUICES		
ZINC		
Average	0.610	0.870
Positive composites		
Total number	9	9
Number reported as trace	0	0
Range	0.300–1.00	0.200–2.40
LEAD		
Average	0.066	0.062
Positive composites		
Total number	5	5
Number reported as trace	0	0
Range	0.070–0.290	0.080–0.240
ENDOSULFAN		
Average	T	
Positive composites		
Total number	1	
Number reported as trace	1	
Range	T	
PERTHANE		
Average	T	
Positive composites		
Total number	1	
Number reported as trace	1	
Range	T	
DICHLORAN		
Average	0.001	0.005
Positive composites		
Total number	2	3
Number reported as trace	0	0
Range	0.006–0.007	0.012–0.026
CADMIUM		
Average	0.007	0.005
Positive composites		
Total number	6	4
Number reported as trace	0	0
Range	0.010–0.017	0.010–0.020
CARBARYL		
Average	T	
Positive composites		
Total number	1	
Number reported as trace	1	
Range	T	
PCP		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
ETHION		
Average	T	T
Positive composites		
Total number	1	1
Number reported as trace	1	1
Range	T	T
o-PHENYLPHENOL		
Average		T
Positive composites		
Total number		1
Number reported as trace		1
Range		T
IX. OILS AND FATS		
ZINC		
Average	20.3	14.43
Positive composites		
Total number	2	10
Number reported as trace	0	0
Range	20.2–20.4	10.6–19.2
CADMIUM		
Average	0.074	0.062
Positive composites		
Total number	2	10
Number reported as trace	0	0
Range	0.067–0.080	0.040–0.110
MALATHION		
Average	0.025	0.041
Positive composites		
Total number	2	6
Number reported as trace	0	0
Range	0.015–0.035	0.005–0.187
TOXAPHENE		
Average	T	0.007
Positive composites		
Total number	2	4
Number reported as trace	2	3
Range	T	0.075
DIELDRIN		
Average	0.001	0.001
Positive composites		
Total number	1	5
Number reported as trace	0	3
Range	0.002	0.002–0.007
ENDRIN		
Average	0.006	0.001
Positive composites		
Total number	2	1
Number reported as trace	1	0
Range	0.011	0.009
HEPTACHLOR EPOXIDE		
Average	T	0.001
Positive composites		
Total number	2	2
Number reported as trace	2	1
Range	T	0.007
FONOFOS		
Average	T	T
Positive composites		
Total number	1	2
Number reported as trace	1	0
Range	T	0.001–0.002
CHLORDANE		
Average	T	0.014
Positive composites		
Total number	1	3
Number reported as trace	1	2
Range	T	0.137
p,p'-DDE		
Average		T
Positive composites		
Total number		1
Number reported as trace		1
Range		T
LEAD		
Average		0.050
Positive composites		
Total number		6
Number reported as trace		0
Range		0.060–0.120
HCB		
Average		0.002
Positive composites		
Total number		4
Number reported as trace		1
Range		0.001–0.016
PCNB		
Average		0.001
Positive composites		
Total number		3
Number reported as trace		0
Range		0.001–0.005
PCA		
Average		0.005
Positive composites		
Total number		4
Number reported as trace		0
Range		0.002–0.044
PENTACHLOROBENZENE		
Average		0.001
Positive composites		
Total number		4
Number reported as trace		1
Range		0.001–0.009

TABLE 4 (cont'd.). *Levels of chemical and metal residues, by food class, in infant and toddler food composites from 10 United States cities—August 1975–July 1976*

Chemical	Residue, ppm	
	Infant	Toddler
SELENIUM		
Average		0.011
Positive composites		
Total number		2
Number reported as trace		0
Range		0.050–0.060
PARATHION		
Average		T
Positive composites		
Total number		2
Number reported as trace		2
Range		T
ARSENIC		
Average		0.003
Positive composites		
Total number		1
Number reported as trace		0
Range		0.030
PCTA		
Average		T
Positive composites		
Total number		1
Number reported as trace		0
Range		0.003
PCP METHYL ETHER		
Average		T
Positive composites		
Total number		1
Number reported as trace		0
Range		0.002
X. SUGAR AND ADJUNCTS		
ZINC		
Average	2.73	5.51
Positive composites		
Total number	7	10
Number reported as trace	0	0
Range	0.20–20.7	1.50–12.5
CADMIUM		
Average	0.005	0.015
Positive composites		
Total number	6	10
Number reported as trace	0	0
Range	0.007–0.010	0.010–0.020
MERCURY		
Average	0.0006	
Positive composites		
Total number	1	
Number reported as trace	0	
Range	0.006	
SELENIUM		
Average		0.004
Positive composites		
Total number		1
Number reported as trace		0
Range		0.040
PCP		
Average		0.007
Positive composites		
Total number		1
Number reported as trace		0
Range		0.070
LINDANE		
Average		0.001
Positive composites		
Total number		6
Number reported as trace		0
Range		0.001–0.003
α-BHC		
Average		T
Positive composites		
Total number		6
Number reported as trace		3
Range		0.001–0.002
LEAD		
Average		0.028
Positive composites		
Total number		2
Number reported as trace		0
Range		0.090–0.190
ARSENIC		
Average		0.004
Positive composites		
Total number		1
Number reported as trace		0
Range		0.040
2,4-D		
Average		0.002
Positive composites		
Total number		1
Number reported as trace		0
Range		0.025
XI. BEVERAGES		
ZINC		
Average	0.314	0.180
Positive composites		
Total number	5	7
Number reported as trace	0	0
Range	0.100–1.00	0.100–0.500
CADMIUM		
Average	0.007	0.002
Positive composites		
Total number	1	2
Number reported as trace	0	0
Range	0.012	0.010
LEAD		
Average	0.011	0.007
Positive composites		
Total number	1	1
Number reported as trace	0	0
Range	0.080	0.070

NOTE: Average residues are based upon the total number of composites examined; trace values were treated as zero. It is quite possible that average values reported as T can be well below the detection limits of the method for that composite.

ppm and 0.045 ppm, respectively, and were found in all 10 composites; lead was found in six composites, averaging 0.078 ppm for the series.

VEGETABLES

Infant—The highest level for a chlorinated pesticide was 0.037 ppm endosulfan found in one composite. There were two reports of *p,p'*-DDE, 0.002 ppm and 0.004 ppm, and trace findings of parathion and *p,p'*-DDT. Average residues of metals included 3.66 ppm zinc, 0.078 ppm lead, 0.043 ppm cadmium, and a trace of mercury.

Toddler—Trace averages were reported for parathion, dieldrin, lindane, PCP, *p,p'*-DDE and α-BHC. Zinc, lead, and cadmium accounted for 26 of the total 36 residues and averaged 3.49, 0.081, and 0.026 ppm, respectively.

FRUITS AND FRUIT JUICES

Infant—The chlorinated pesticide Perthane® and the carbamate pesticide carbaryl were found only in this food group, in one composite at the trace level. Other

TABLE 5. *Intake of pesticide and industrial chemical residues by infants and toddlers, market basket surveys FY 1975 vs FY 1976*

Compound	Residue, µg/kg Body Wt/Day			
	Infant		Toddler	
	FY 75	FY 76	FY 75	FY 76
Aldrin	ND	ND	ND	ND
Dieldrin	0.0153	0.0249	0.0502	0.0412
TOTAL	0.0153	0.0249	0.0502	0.0412
DDE	0.1276	0.0682	0.1598	0.0985
DDT	T	T	0.0064	0.0046
TDE	T	ND	0.0037	0.0018
TOTAL	0.1276	0.0682	0.1699	0.1049
Heptachlor epoxide	0.0097	0.0001	0.0057	0.0057
Hepachlor	ND	ND	ND	ND
TOTAL	0.0097	0.0001	0.0057	0.0057
Endosulfan I	ND	0.0011	ND	ND
Endosulfan II	ND	0.0045	ND	ND
Endosulfan sulfate	ND	0.0368	0.0078	ND
TOTAL	ND	0.0424	0.0078	ND
BHC	0.0228	0.0055	0.0211	0.0132
Captan	ND	ND	ND	ND
Carbaryl	T	T	0.0047	ND
Chlordane	ND	T	ND	0.0100
CIPC	0.0020	0.0321	0.0458	0.3942
2,4–D	ND	ND	ND	0.0058
DCPA	ND	ND	ND	ND
Diazinon	T	0.0053	0.0067	0.0030
Dichloran	0.0383	0.0230	0.0506	0.0342
Dicofol	ND	ND	ND	ND
Ethion	ND	T	ND	ND
Endrin	ND	ND	ND	0.0007
Fonofos	ND	T	ND	0.0002
HCB	0.0044	0.0009	0.0064	0.0042
Leptophos	ND	ND	0.0033	ND
Lindane	0.0133	0.0049	0.0341	0.0096
Malathion	0.2028	0.0865	0.1374	0.1488
Methoxychlor	T	T	ND	T
Methyl parathion	ND	ND	ND	ND
o-Phenylphenol	ND	ND	ND	T
Oxychlordane	T	0.0008	T	T
Parathion	T	T	ND	0.0013
PCA	0.0053	ND	0.0058	0.0064
Pentachloroanisole	ND	ND	ND	0.0003
Pentachlorobenzene	0.0005	ND	0.0007	0.0013
PCNB	0.0073	ND	0.0024	0.0007
PCP	0.0154	T	0.0214	0.0162
PCTA (Pentachlorothioanisole)	ND	ND	ND	0.0004
Perthane	ND	T	ND	ND
Phosalone	ND	ND	ND	ND
PCBs	T	T	T	ND
Ronnel	ND	ND	T	0.0022
Toxaphene	0.2573	T	0.0467	0.0127
trans-Nonachlor	ND	ND	ND	0.0050
TCNB	ND	0.0019	T	0.0074
Carbophenothion	ND	ND	ND	ND

NOTE: ND = not detected; T = Trace (below the limits of quantitation; detected and verified, but not quantifiable).

pesticides reported at the trace level included endosulfan, PCP, and ethion. Dichloran was found in two composites and averaged 0.001 ppm for the series. Low-level metal residues included zinc, lead, and cadmium, averaging 0.610, 0.066, and 0.007 ppm, respectively.

Toddler—Dichloran, found in three composites, ranged from 0.012 to 0.026 ppm for a series average of 0.005 ppm. Traces of PCP, ethion, and the fungicide *o*-phenylphenol were also reported. Zinc, lead, and cadmium all averaged below 1.0 ppm.

TABLE 6. *Dietary intakes of metals by infants and toddlers, FY 1975 vs FY 1976*

Metal	Residue, µg/day			
	Infant		Toddler	
	FY 75	FY 76	FY 75	FY 76
Lead	20.79	26.94	25.61	30.12
Cadmium	5.16	12.33	10.72	14.19
Zinc [1]	5.33	8.15	8.26	9.46
Arsenic [2]	2.76	0.42	11.10	12.28
Selenium	21.63	10.81	58.38	44.99
Mercury	0.03	0.56	0.94	0.81

[1] Values are mg/day.
[2] Values calculated as arsenic trioxide (As_2O_3).

OILS AND FATS

Infant—Only two of the 10 market baskets included a separate composite of oils and fats for the infant. Malathion, endrin, and dieldrin averaged 0.025, 0.006, and and 0.001 ppm, respectively. Trace averages were reported for toxaphene, heptachlor epoxide, and chlordane.

Fonofos, an organophosphorus pesticide, was found only in this food group, at the trace level. Both composites had residues of zinc, 20.2 ppm and 20.4 ppm, and cadmium, 0.067 ppm and 0.080 ppm.

Toddler—A total of 71 residues was reported for these 10 composites. The metals accounted for 29 residues and included zinc (average 14.43 ppm), cadmium (average 0.062 ppm), lead, selenium, and arsenic. Three organophosphorus pesticides were reported: malathion in six composites, ranging from 0.005 to 0.187 ppm and averaging 0.041 ppm for the series; parathion traces in two composites; and fonofos in two composites, ranging from 0.001 to 0.002 ppm and averaging trace for the series. Malathion residues averaged highest of the nonmetals and appeared in the most composites. Among the organochlorine compounds, toxaphene averaged 0.007 ppm and was reported in four composites. Other organochlorine residues included PCA, dieldrin, pentachlorobenzene, chlordane (average 0.014 ppm), HCB, heptachlor epoxide, PCNB, *p,p'*-DDE, PCTA, and PCP methyl ether.

SUGAR AND ADJUNCTS

Infant—Only three metal residues were reported for these infant composites. Zinc was found in seven composites, ranging from 0.20 to 20.7 ppm, for a series average of 2.73 ppm. Cadmium was found in six com-

posites, ranging from 0.007 to 0.010 ppm and averaging 0.005 ppm for the series. One residue of mercury was reported at 0.006 ppm for a series average of 0.0006 ppm.

Toddler—In contrast, the toddler composites had 38 residues of nine different compounds: five metals, two chlorinated herbicides, and two organochlorine pesticides. Averages of the metals included 5.51 ppm zinc, 0.015 ppm cadmium, 0.028 ppm lead, and 0.004 ppm each for arsenic and selenium. Two composites contained herbicides: PCP at 0.07 ppm and 2,4-D at 0.025 ppm. Six residues were reported for each of the chlorinated pesticides, lindane and α-BHC, with ranges 0.001 to 0.003 ppm and 0.001 to 0.002 ppm, respectively.

BEVERAGES

Infant—Only three metal residues were reported for these composites. Zinc was found in five composites, ranging 0.100 to 1.00 ppm, and averaging 0.314 ppm for the series; 0.012 ppm cadmium and 0.080 ppm lead were reported for one composite.

Toddler—The metal residues found in these toddler composites were the same as those found in the infant composites. Zinc was found in seven composites, ranging from 0.100 to 0.500 ppm, and averaging 0.180 ppm for the series. Cadmium was reported twice at the 0.01 ppm level, and lead was reported once at 0.070 ppm.

Discussion

INFANT

The infant composites contained a total of 301 residues: 207, or 68.7 percent, were metals; 86, or 28.6 percent, were pesticides; and the remaining 2.7 percent included seven herbicide residues and one fungicide residue. In comparison, a total of 306 residues was reported for the first infant composite series: 199 (65 percent) metals, 99 (32.3 percent) pesticides, five fungicides, one herbicide, and two industrial chemicals.

Of the metal residues, zinc, occurring most frequently, was found in 85 composites with the highest level, 36.3 ppm, occurring in the meat-fish-poultry composite. Although 59 cadmium residues were reported, the 35 lead residues, ranging from 0.050 to 0.290 ppm, might be of greater significance. The highest level of lead residues was found in the fruit and fruit juice composites. All but one of the 21 selenium residues were found in the meat-fish-poultry composites. Arsenic was reported at low levels in three grain-cereal composites. Sixteen chlorinated pesticide compounds were reported in 69 residues; 43 were found at the trace level. Of these, the most frequently occurring residues were dieldrin and *p,p'*-DDE, found mostly in the dairy composites and the meat composites. Endosulfan, found in one vegetable composite at 0.037 ppm, was the chlorinated pesticide occurring at the highest level.

Malathion, an organophosphorus pesticide, was found in each of the 10 grain and cereal composites, with a high value of 0.049 ppm. In addition to 0.010 ppm diazinon found in one composite, single trace amounts of three other organophosphorus pesticides were reported: parathion, ethion, and fonofos. A single trace of carbaryl, the only carbamate pesticide screened, was found once in the fruit-fruit juice composite.

Only two herbicides were found in the composites. The chlorinated herbicide CIPC, which is usually found in potatoes, was reported for three potato composites, ranging from 0.023 to 0.469 ppm, and trace amounts of PCP were found in four composites. The fungicide TCNB was reported in one potato composite at 0.026 ppm.

TODDLER

A total of 473 residues was found in the toddler food composites. Of these, the six metals accounted for 257 residues, or 54.3 percent of the total, the 17 chlorinated pesticides accounted for 164, or 34.6 percent, and the six organophosphorus pesticides were reported 26 times for 5.4 percent of the total. The remaining 5.7 percent included four chlorinated herbicides found 13 times, three chlorinated fungicides found eight times, one industrial chemical found four times, and *o*-phenylphenol found once.

Zinc, ranging from 0.100 to 34.0 ppm, was found in almost every composite. Cadmium was the second most frequently occurring residue, but the range, 0.005 to 0.120 ppm, was much lower. Most of the lead residues, with a range of 0.050 to 0.350 ppm, were found in the grain, potato, vegetable, and fruit composites. All 10 grain composites and nine meat composites contained selenium residues. The meat-fish-poultry composites contained seven of the 11 arsenic residues and six of the seven mercury residues, which have been traced to the seafood portion of the composite.

Seventeen chlorinated pesticides accounted for 164, or 34.6 percent, of all the residues; these were found predominantly in the dairy and meat-fish-poultry composites. The most frequently detected chlorinated pesticide was dieldrin, found in every dairy and meat-fish-poultry composite. α-BHC and DDE were also found several times in those two composites.

The most prevalent organophosphorus pesticide was malathion, found in all 10 grain-cereal composites and in six oil-fat composites. Other reportable organophosphorus pesticides were diazinon, parathion, fonofos, and ronnel, each residue found in either the grain-cereal or the oil-fat composites. The herbicides and

fungicides represented a small number of residues. Only four chlorinated herbicides were reported: CIPC residues, found in five potato composites, ranged from 0.023 to 0.469 ppm; PCP, 2,4-D, and PCP methyl ether were scattered throughout the other food groups. Most of the eight residues of the three chlorinated fungicides detected were found in the fat-oil composite.

Pentachlorobenzene, an industrial chemical, was present in the toddler diet but was not found in the infant diet; it was detected in four oil-fat composites and ranged from 0.001 to 0.009 ppm. The fruit-fruit juice composites contained one residue of the fungicide *o*-phenylphenol at the trace level.

Table 7 shows the number of occurrences of each residue type as found in each food class.

Recovery studies were done for many of the more common residues. In each case, simultaneous determinations were made on an unfortified composite and on a composite fortified with a known level of residue. Table 8 lists the contributions from the unfortified composite, and the total amount of residue recovered through the method. A single determination was made for each reported recovery. In some cases only a few composites were fortified with a particular compound; these are presented for information only. In other cases, an attempt was made to investigate the recovery of the most frequently found residues from a variety of product matrices and to provide a basis for a meaningful evaluation of the methods. The data are presented in Table 8.

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TABLE 7. *Types and number of residues, by food class, found in infant and toddler total diet samples from 10 United States cities—August 1975–July 1976*

Type of Residue	Food Class [1] I	II	III	IV	V	VI	VII	VIII	IX	X	XI
INFANT											
Metals	2	3	3	5	5	3	4	3	2	3	3
Pesticides	—	4	5	5	3	6	4	5	7	—	—
Fungicides	—	—	—	—	—	2	—	—	—	—	—
Industrial chemicals	—	—	—	—	—	—	—	—	—	—	—
Herbicides	—	1	1	—	1	—	—	1	—	—	—
TODDLER											
Metals	2	3	5	6	5	3	3	3	5	5	3
Pesticides	—	4	8	11	2	5	5	2	11	2	—
Fungicides	—	—	—	—	—	2	—	1	2	—	—
Industrial chemicals	—	—	—	—	—	—	—	—	1	—	—
Herbicides	—	1	1	—	1	—	1	1	1	2	—

[1] See key in Table 1.

TABLE 8. *Recovery data on residues found in infant and toddler total diet samples from 10 United States cities—August 1975–July 1976*

Residue	Type of Food Composite	Spike Level, ppm	Range of Unfortified Composite, ppm [1]	Range of Total Residue Found, ppm [1, 2]	Number of Recovery Studies
NONMETALS					
Oxychlordane	Fatty	0.003		0.0027–0.0039	2
	Nonfatty	0.003		0.0026	1
Heptachlor epoxide	Fatty	0.003		0.0027–0.0031	2
	Nonfatty	0.003		0.0027	1
Ethion	Fatty	0.010		0 –trace	2
	Nonfatty	0.010		0.010	1
DCPA	Fatty	0.005	0–0.001	0.0046–0.007	4
	Nonfatty	0.005		0.00043–0.0064 (0.0043)	8
Methyl parathion	Fatty	0.005		0.0011–0.003	2
	Nonfatty	0.005		0.0014–0.0053	4
Perthane	Fatty	0.010		0.005–0.0094	3
	Nonfatty	0.010		0.0009–0.0131 (0.0087)	8
Tetradifon	Fatty	0.100		0.065–0.090	2
	Fatty	0.020		0.019	1
	Nonfatty	0.100		0.048–0.109	4
	Nonfatty	0.020		0.016–0.024	2
Endosulfan sulfate	Fatty	0.010		0.006–0.008	2
	Nonfatty	0.010		0.004–0.013	4
Malathion	Fatty	0.005		0.0025–0.0049	2
	Nonfatty	0.005		0.0043–0.0056	3
Phosalone	Fatty	0.02		0.018	1
	Nonfatty	0.02		0.009–0.015	2
Leptofos	Fatty	0.05		0.039–0.049	2
	Nonfatty	0.05		0.004–0.046	4
PCP	Fatty	0.02	0–0.010	0.0007–0.018	4
	Fatty	0.04		0.020–0.024	3
	Nonfatty	0.02	0–0.003 (0.0012)	0.003–0.016 (0.0096)	6
	Nonfatty	0.04	0–0.004	0.025–0.032	3
Picloram	Fatty	0.10		0–0.075	2
	Nonfatty	0.10		0.033	1
2,4–D	Fatty	0.04		0–0.042	3
	Nonfatty	0.04		0.026–0.039	5
Silvex	Fatty	0.04		0.008–0.022	2
	Nonfatty	0.04		0.023–0.039	2
MCP 2-methyl-4-chloro-phenoxy-acetic acid	Fatty	0.020		0.009–0.011	2
	Nonfatty	0.020		0.009–0.013	3
	Nonfatty	0.040		0.029–0.036	2
2,4–DB	Fatty	0.02		0.014	1
	Nonfatty	0.02		0.012–0.017	2
	Nonfatty	0.04		0–0.026	2
2,4,5–T	Fatty	0.02	0–0.002	0.004–0.022	2
	Fatty	0.04		0.016	1
	Nonfatty	0.02		0.007–0.0196	4
	Nonfatty	0.04		0.025–0.029	2
Carbaryl	Nonfatty	0.20		0.02–0.20 (0.18)	17
o-Phenyl-phenol	Nonfatty	0.40		0.08–0.40 (0.30)	16
Fonofos	Fatty	0.01		0.002–0.004	2
	Nonfatty	0.01		0.006–0.008 (0.007)	5
Toxaphene	Fatty	0.20		0.201–0.240	2
	Nonfatty	0.20		0.125–0.204 (0.170)	4
METALS					
Arsenic	Fatty	0.30		0.25–0.30	3
	Fatty	0.40	0–0.10	0.34–0.535 (0.462)	5
	Nonfatty	0.30		0.17–0.32	3
	Nonfatty	0.40	0–0.03	0.24–0.385 (0.313)	12
Cadmium	Fatty	0.10	0.002–0.086 (0.030)	0.095–0.162 (0.116)	11
	Nonfatty	0.10	0–0.371 (0.0195)	0.090–0.162 (0.116)	19
Lead	Fatty	0.20	0–0.119 (0.039)	0.099–0.260 (0.187)	11
	Nonfatty	0.20	0–0.072 (0.031)	0.084–0.345 (0.196)	19

TABLE 8 (cont'd.). *Recovery data on residues found in infant and toddler total diet samples from 10 Unted States cities—August 1975–July 1976*

Residue	Type of Food Composite	Spike Level, ppm	Range of Unfortified Composite, ppm [1]	Range of Total Residue Found, ppm [1, 2]	Number of Recovery Studies
Mercury	Fatty	0.06	0–0.019 (0.005)	0.047–0.089 (0.069)	7
	Nonfatty	0.04	0–0.003 (0.001)	0.045–0.056 (0.052)	4
	Nonfatty	0.06	0–0.002 (0.0002)	0.065–0.136 (0.079)	13
Selenium	Fatty	0.20	0–0.20 (0.07)	0.19–0.45 (0.30)	6
	Fatty	0.40	0–0.20 (0.03)	0.26–0.50 (0.39)	5
	Nonfatty	0.20	0–0.29 (0.04)	0.18–0.56 (0.25)	11
	Nonfatty	0.40	0–0.15 (0.034)	0.25–0.54 (0.36)	6
Zinc	Fatty	5.0	1.62–19.2 (7.33)	5.95–24.0 (12.2)	6
	Fatty	25.0	5.0–30.2 (17.9)	28.5–52.6 (41.7)	5
	Nonfatty	10.0	6.3–7.4 (7.0)	14.8–18.2 (16.8)	3
	Nonfatty	5.0	0–7.55 (2.67)	4.55–12.6 (7.37)	14

[1] Numbers in parentheses represent average residue levels.
[2] Values are uncorrected for background.

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Organochlorine Pesticides and PCBs in Cod-Liver Oil of Baltic Origin, 1971–80

Jerzy Falandysz [1]

ABSTRACT

Organochlorine pesticides and polychlorinated biphenyls (PCBs) in cod-liver oil of Baltic origin were monitored during 1971–80. Residues of DDT and its metabolites, PCBs, and hexachlorobenzene were present in all samples. Generally, ΣDDT residues declined, but the reason for the decline is unknown.

Introduction

Use of technical DDT as an insecticide in countries surrounding the Baltic has been prohibited since 1971. The organochlorine pesticides presently used for agricultural purposes in Poland are toxaphene, methoxychlor, endosulfan, and lindane. DDT was widely used in agriculture and has been detected at high levels in tissues of Baltic marine mammals, fish, and birds (*7*). The prohibition on the agricultural use of DDT in Poland was reflected in the nearly tenfold reduction of ΣDDT residue levels in adipose fat of slaughtered animals during the last decade (A. Niewiadowska, Veterinary Institute, Pulawy, 1980, personal communication). However, so far there have been no reports showing a decline of ΣDDT residue levels in animals of the Baltic Sea. Another class of organochlorine compounds, polychlorinated biphenyls (PCBs), occurs throughout the Baltic environment. Also, polychlorinated terphenyls (PCTs) have been found recently in Baltic marine organisms (*2, 13, 15*). Information on production and industrial use of PCBs and PCTS in countries surrounding the Baltic remains obscure. Some quantities of PCBs are produced and available in Poland under the trade name Chlorofen (*11, 12*). PCBs have also been produced in West Germany as Clophen, in the Soviet Union as Sovol, and in Czechoslovakia as Delor. This paper presents the results of analyses of cod-liver oil of Baltic origin for residues of hexachlorobenzene (HCB), ΣDDT, and PCBs.

Analytical Methods

Samples of cod-liver oil were obtained from a factory in Gdynia. The procedures for isolation and determination of the organochlorines have been presented elsewhere (*4*). Two cleanup processes were used. The oil sample was dissolved in *n*-hexane; 1 ml *n*-hexane was used for every 20 mg fat extracted. Twenty mg fat was cleaned with 1 ml of a 1:1 mixture of fuming 20–25 percent sulfur dioxide and concentrated sulfuric acid in a screw-capped, Teflon-lined test tube (*2, 4, 8*). The colorless hexane layer was injected directly into the gas chromatograph. In addition, a 4-ml aliquot of cleaned sample was subjected to alcoholic potassium hydroxide hydrolysis in a screw-capped, Teflon-lined test tube. The test tube was immersed for 15 minutes in the water bath at 50°C. After cooling, the mixture was shaken with 4 ml distilled water. Following separation, the upper hexane layer was injected into the gas chromatograph. The residues were quantitated by electron-capture gas chromatography. Instrument parameters and operating conditions follow:

Chromatograph:	PYE 104
Detector:	^{63}Ni
Column:	glass, 150 cm long by 4 mm ID packed with a 2:1 mixture of 8 percent QF-1 and 4 percent SF-96 on 100–120 mesh Gas-Chrom Q
Temperatures, °C:	detector 210
	column oven 195
Carrier gas:	argon flowing at ml/min

PCBs were quantified by comparing sample peaks with that of PCB standard Clophen A 50, which appeared on the gas chromatogram after *p,p'*-DDE. DDE content was calculated from the total height of the peak with retention time equal to that of standard *p,p'*-DDE. *p,p'*-DDT and *p,p'*-TDE were calculated from the difference in height of peaks with retention times equal to those of *p,p'*-DDT and *p,p'*-TDE before and after hydrolysis with alcoholic potassium hydroxide. In recovery experiments, the calculated values and standard deviations, in ppm, were as follows: HCB, 0.27 ± 0.015; DDE, 5.8 ± 0.25; TDE, 5.1 ± 0.39; DDT, 1.3 ± 0.14; and PCBs, 11 ± 0.84 (*4*).

Results and Discussion

The levels of organochlorine residues in cod-liver oil are presented in Table 1. DDT and its metabolites were present in all samples. DDE levels ranged from 1.1 to

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TABLE 1. *Mean and range (ppm) of chlorinated hydrocarbons in cod-liver oil of Baltic origin, 1971–80*

Year	n	HCB		p,p′-DDE		p,p′-TDE		p,p′-DDT		ΣDDT		PCBs	
		Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
1971	1	0.24		4.6		2.3		6.6		13		4.8	
1972	1	0.36		4.2		4.6		8.4		17		9.5	
1973	3	0.28	0.20–0.32	7.2	4.1–9.3	3.7	2.2–4.5	10	8.4–12	22	15–26	5.7	4.3–7.0
1974	8	0.23	0.10–0.37	5.5	1.3–9.8	2.9	0.69–4.8	5.7	2.1–9.4	14	4.0–22	5.9	2.2–9.0
1975	5	0.23	0.12–0.34	8.3	2.9–17	3.5	1.4–5.4	5.0	2.2–8.5	17	8.9–28	8.6	4.6–13
1976	11	0.30	0.16–0.44	8.3	1.1–24	3.5	1.0–6.5	4.6	1.1–8.9	16	3.3–38	8.6	3.1–16
1977	9	0.35	0.26–0.54	5.3	3.1–9.3	3.9	3.0–5.0	3.9	3.2–5.9	13	9.8–20	9.6	4.9–16
1978	4	0.32	0.28–0.41	4.1	4.1–4.2	2.9	2.7–3.1	2.2	1.3–3.0	9.2	8.2–9.8	7.1	5.5–8.5
1979	1	0.24		4.2		5.2		2.1		11		4.3	
1980	2	0.29	0.28–0.30	3.4	3.0–3.8	1.9	1.3–2.5	1.2	0.59–1.8	6.5	4.9–8.1	5.1	4.7–5.6

NOTE: Mean is arithmetic mean.

24 ppm, TDE from 0.69 to 6.5 ppm, and DDT from 0.59 to 12 ppm; ΣDDT levels ranged from 3.3 to 38 ppm. PCBs, resembling Clophen A 50, and HCB were also detected in all samples, at levels ranging from 2.2 to 16 ppm and from 0.10 to 0.54 ppm, respectively. All samples contained at least trace amounts of α- and γ-BHC. Because the residues of these isomers were negligible by comparison, they were excluded from the table.

Many papers have been published on organochlorine residues in the liver of cod from the Baltic (*1, 5, 6, 8, 10, 11, 13, 14, 16-18*), but only several for cod-liver oil (*1, 3, 10*). The cod-liver oil of Baltic origin is unfit for medical purposes because of its high contamination with DDT and PCBs.The main Polish catches of cod are taken in the southern Baltic (Figure 1, regions 25 and 26) (*9*). The cod-liver oil produced in the factory in Gdynia, Poland, is manufactured from fresh cod livers. The crude cod-liver oil produced on board the fishing vessels operating in the southern Baltic is also delivered to the factory in Gdynia. After clarification, the crude cod-liver oil is usually mixed with that produced from the previous catch and is stored in a large-capacity tank. For the present study, all samples were taken from oil dispatched from the factory. Those particular lots of oil were taken from the day-by-day production or from the storage tank, mainly mixed oil. The annual production of cod-liver oil in 1975, 1976, and 1977 was 280, 316, and 220 tons, respectively. Table 1 shows that ΣDDT may have declined, but that decline cannot be verified because the mean values for organochlorines must be at least corrected by the quantity of oil in the particular lot analyzed—information that was not available to authors. Also, it has been shown (*11, 16, 17*) that the organochlorine residues in livers of cod from the western Baltic can be correlated to the length of the fish. The lengths of the cod from which livers were obtained for processing in the present study were not uniform. Cod caught in the southern Baltic generally range from 25 to 120 cm long. In 1970, the cod were predominantly 39 to 44 cm long, and in 1971, 45 to 50 cm long (*9*).

It should be pointed out that much higher levels of organochlorines than those noted in Table 1 have been found in the livers of some specimens of cod from the Baltic. For example, the livers of cod taken from the Kattegat in 1973 contained PCBs ranging from 4 to 53 ppm and averaging 13 ppm wet weight; livers of cod taken from the Rivo Fiord in 1975 contained PCBs ranging from 29 to 57 ppm and averaging 38 ppm wet weight (*13, 14*).

The following extreme ranges for ΣDDT have also been noted: Levels in cod taken from the southern Baltic in 1970–71 ranged from 14 to 57 ppm and averaged 26 ppm wet weight and 66 ppm lipid weight (*6*); levels in cod taken from the Sund during 1973 ranged from 1.4 to 53 ppm and averaged 8.7 ppm wet weight and 29 ppm lipid weight (*13*).

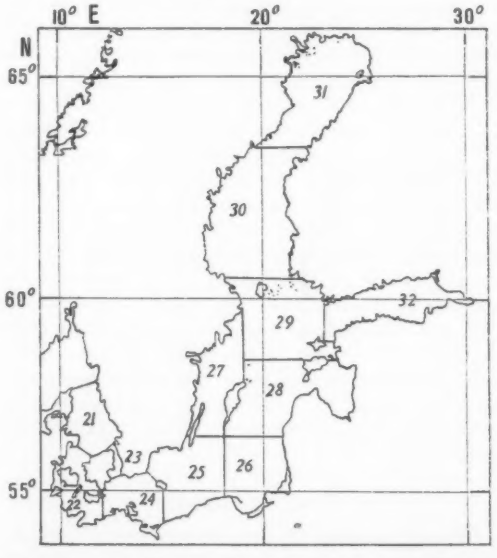

FIGURE 1. *Baltic Sea, with division of the sampling areas according to International Council for the Exploration of the Sea*

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Pesticide, Metal, and Other Chemical Residues in Adult Total Diet Samples—(XII)—August 1975–July 1976 [1]

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ABSTRACT

This report is the twelfth in the series on the presence of pesticide and other chemical residues in the average diet of the United States' heartiest eater, the young adult male. Twenty market baskets were collected in 20 U.S. cities that ranged in population from < 50,000 to 1 million or more. Composites of 12 food classes were analyzed. Averages and ranges of residues found are reported for August 1975 through July 1976, by food class. In addition to the pesticide and chemical residues, data for lead, cadmium, selenium, mercury, arsenic, and zinc are included. The individual items making up the dairy and meat composites in four market baskets were analyzed separately for pesticide residues, and the results are included. Results of recovery studies of pesticides and chemicals within various food classes are also presented.

Introduction

In 1964, the Food and Drug Administration (FDA), U.S. Department of Health and Human Services (formerly U.S. Department of Health, Education, and Welfare), initiated a Total Diet Program (*7*), sometimes called the Market Basket study. Its purpose was to monitor the atmosphere for fission products from atmospheric tests of thermonuclear weapons in May 1961. Later, the program was expanded to include pesticide residues and certain nutrients.

At its inception, the program was primarily concerned with the adult diet, which was defined as a market basket of food representing the basic two-week diet of a 16-to-19-year-old male, statistically the United States' heartiest eater. Beginning in August 1974, 10 of the 30 market baskets collected per year were changed to represent the basic two-week diet of infants (6-month-old) and toddlers (2-year-old) (*13*).

The market baskets were collected in four different geographic areas, with the specific diet of the particular region determining the composition of the market basket. Foods were prepared for normal home consumption, and every food item was then placed into one of the 12 composite classes listed in Table 1. For every food class, 20 composites, one from each market basket, were prepared. Each composite, containing foods with similar characteristics, was analyzed for certain metals, residues of organochlorine, organophosphorus, and carbamate pesticides, herbicides, and industrial chemicals. Methodologies included atomic absorption spectroscopy, fluorometry, polarography, gas chromatography, thin-layer chromatography, mass spectroscopy, and established extraction and cleanup techniques (*8–10, 18, 19*). Amounts and types of residues found from June 1964 through July 1975 have been tabulated in earlier reports (*1–5, 11–17*). This report covers the results obtained from August 1975 through July 1976 for adult market baskets collected in 20 different cities. Results for the 10 infant and toddler market baskets collected during the same period are presented in a separate report.

Results

During this reporting period, 1,039 residues of 47 different compounds were found in the 240 composites examined. In the previous reporting period, 959 residues of 42 different compounds were found in the same number of composites. The 47 compounds are listed in decreasing order of frequency in Table 2. Table 3 shows the frequency of occurrence of each compound by food class, and Table 4 shows the levels of every residue found within each food class. The average value in Table

TABLE 1. *Classes of adult food composites analyzed for pesticides, metals, and other chemical residues, August 1975–July 1976*

Key	Food Class
I	Dairy products
II	Meat, fish, and poultry
III	Grain and cereal products
IV	Potatoes
V	Leafy vegetables
VI	Legume vegetables
VII	Root vegetables
VIII	Garden fruits
IX	Fruits
X	Oils, fats, and shortening
XI	Sugar and adjuncts
XII	Beverages (including drinking water)

[1] Food and Drug Administration, Kansas City District Office Laboratory, 1009 Cherry St., Kansas City, MO 64106

TABLE 2. *Chemical and metal residues found in adult food composites from 20 United States cities—August 1975–July 1976*

Chemical Found	No. of Composites with Residues	No. of Positive Composites with Residues Reported as Trace [1]	Range, ppm
Zinc	239	0	0.100 –76.0
Cadmium	170	0	0.010 – 0.100
Lead	85	0	0.040 – 0.820
Dieldrin	60	16	0.001 – 0.086
Selenium	57	0	0.010 – 0.340
p,p'-DDE [2]	52	16	0.001 – 0.048
α-BHC	46	31	0.0003– 0.007
Arsenic	31	0	0.030 – 0.460
Heptachlor epoxide	30	19	0.001 – 0.003
Malathion	29	3	0.004 – 0.096
Mercury	24	0	0.006 – 0.080
Lindane	24	11	0.0006– 0.004
HCB	19	12	0.0002– 0.0060
Octachlor epoxide	17	15	0.0020
p,p'-DDT	16	9	0.0030– 0.010
Endosulfan sulfate	13	5	0.003 – 0.030
Endosulfan I	12	1	0.0010– 0.0110
Endosulfan II	11	5	0.002 – 0.0120
Dichloran	10	1	0.002 – 0.163
Diazinon	10	2	0.001 – 0.004
TCNB	8	0	0.002 – 0.114
p,p'-TDE	7	6	0.004
Ethion	6	2	0.005 – 0.050
Parathion	5	0	0.002 – 0.006
CIPC	5	0	0.010 – 0.229
Dacthal®	5	1	0.002 – 0.014
PCNB	5	2	0.002 – 0.003
Carbaryl	5	4	0.050
Dicofol	4	0	0.007 – 0.028
Pentachloroaniline	4	1	0.007 – 0.018
Perthane	3	0	0.014 – 0.044
Methoxychlor	3	2	0.013
PCB	3	3	T
Captan	2	0	0.026 – 0.040
PCP	2	0	0.026 – 0.060
Pentachlorobenzene	2	0	0.004 – 0.005
PCTA	2	0	0.002 – 0.005
Ronnel	2	0	0.002 – 0.006
Leptophos	2	1	0.009
Chlordane	2	2	T
β-BHC	1	0	0.008
PCP Methyl ether	1	0	0.002
trans-Nonachlor	1	0	0.002
Carbophenothion	1	0	0.195
Phosalone	1	0	0.008
o-Phenylphenol	1	1	T
Toxaphene	1	1	T

[1] Chemicals capable of being detected by the specific analytical methodology may be confirmed qualitatively but are not quantifiable when they are present at concentrations below the limit of quantitation. Limit of quantitation varies with residue and food class.

TABLE 3. *Frequency of occurrence, by food class, of pesticides, metals, and other chemical residues in adult food composites from 20 United States cities—August 1975–July 1976*

Chemical	Food Class [1] I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
	Number of Occurrences											
Zinc	20	20	20	20	20	20	20	20	20	20	20	19
Cadmium	3	17	20	20	19	14	19	18	5	18	14	3
Lead	0	5	12	5	2	17	8	14	11	6	3	2
Dieldrin	15	19	1	4	2	0	1	13	2	3	0	0
Selenium	4	20	20	3	1	5	2	0	0	2	0	0
p,p'-DDE	14	20	0	3	8	1	3	2	0	1	0	0
α-BHC	17	19	0	0	0	0	0	3	1	1	5	0
Arsenic	1	17	8	0	0	1	1	1	0	1	0	1
Heptachlor epoxide	13	15	0	2	0	0	0	0	0	0	0	0
Malathion	0	0	19	0	0	0	0	0	0	7	3	0
Mercury	0	18	1	0	0	0	0	0	1	1	1	2
Lindane	2	7	1	0	1	0	0	5	0	1	7	0
HCB	5	11	0	0	0	0	0	0	0	3	0	0
Octachlor epoxide	5	12	0	0	0	0	0	0	0	0	0	0
p,p'-DDT	0	15	0	0	1	0	0	0	0	0	0	0
Endosulfan sulfate	0	0	0	2	6	0	0	3	2	0	0	0
Endosulfan I	0	0	0	0	5	0	0	6	1	0	0	0
Endosulfan II	0	0	0	0	5	0	0	4	2	0	0	0
Dichloran	0	0	0	0	4	1	0	0	4	0	1	0
Diazinon	0	0	5	0	2	0	0	2	1	0	0	0
TCNB	0	0	1	6	0	0	0	1	0	0	0	0
p,p'-TDE	0	7	0	0	0	0	0	0	0	0	0	0
Ethion	0	1	0	0	0	0	1	1	3	0	0	0
Parathion	0	0	0	0	1	2	1	1	0	0	0	0
CIPC	0	0	0	5	0	0	0	0	0	0	0	0
DCPA	0	0	0	0	3	0	2	0	0	0	0	0
PCNB	0	0	0	0	0	0	0	0	0	5	0	0
Carbaryl	0	0	0	0	0	0	0	2	3	0	0	0
Dicofol	0	0	0	0	0	0	0	0	4	0	0	0
Pentachloroaniline	0	0	0	0	0	0	0	0	0	4	0	0
Perthane	0	0	0	0	2	0	0	0	1	0	0	0
Methoxychlor	3	0	0	0	0	0	0	0	0	0	0	0
PCB	1	2	0	0	0	0	0	0	0	0	0	0
Captan	0	0	0	0	0	0	0	0	2	0	0	0
PCP	0	0	0	0	0	0	0	0	0	0	1	1
Pentachlorobenzene	0	0	0	0	0	0	0	0	0	2	0	0
PCTA	0	0	0	0	0	0	0	0	0	2	0	0
Ronnel	0	1	1	0	0	0	0	0	0	0	0	0
Leptophos	0	0	0	0	0	0	0	2	0	0	0	0
Chlordane	0	0	1	1	0	0	0	0	0	0	0	0
β-BHC	0	0	0	0	1	0	0	0	0	0	0	0
PCP Methyl ether	0	0	0	0	0	0	0	0	0	1	0	0
trans-Nonachlor	0	1	0	0	0	0	0	0	0	0	0	0
Carbophenothion	0	0	0	0	0	0	0	1	0	0	0	0
Phosalone	0	0	0	0	0	0	0	0	1	0	0	0
o-Phenylphenol	0	0	0	0	0	0	0	0	0	0	1	0
Toxaphene	0	0	0	0	1	0	0	0	0	0	0	0

[1] See Table 1 for key to food classes.

4 is based on the 20 composites examined; trace residues, if present, were treated as zero in calculating the averages. For this reason, an average value reported as "T" can be well below the detection limits of the methods for that compound.

Duggan et al. reported the human dietary intake of pesticides and industrial chemicals detected in mg/kg body weight/day, for the period July 1969 through June 1976 (*6*). Comparative values for fiscal years 1971–75 are also given (*6*). Because Duggan et al. do not report dietary intakes for metals determined in the Total Diet studies, they are shown here in Table 5 for FY 76. The daily intakes in μg/day (mg/day for zinc) are listed in Table 5 by food group, together with the percentage of the total daily intake contributed by each.

The most common residues and their maximum levels for each of the 12 food classes are discussed below. No findings have been corrected for recovery.

DAIRY PRODUCTS

Metal residues were found most frequently and at the highest levels in dairy products. Averages were 4.92 ppm zinc, 0.004 ppm selenium, 0.004 ppm arsenic, and 0.002 ppm cadmium. Of the organochlorine residues, *p,p'*-DDE levels, ranging from 0.001 to 0.010 ppm and averaging 0.002 ppm for the series, were the highest. Other

TABLE 4. *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

Chemical	Residues, ppm	Chemical	Residues, ppm
I. DAIRY PRODUCTS			
ZINC		α-BHC	
Average	4.92	Average	T
Positive composites		Positive composites	
Total number	20	Total number	17
Number reported as trace	0	Number reported as trace	8
Range	3.50–5.90	Range	0.0003–0.0010
p,p'-DDE		METHOXYCHLOR	
Average	0.002	Average	T
Positive composites		Positive composites	
Total number	14	Total number	3
Number reported as trace	6	Number reported as trace	2
Range	0.0010–0.0100	Range	0.013
DIELDRIN		HCB	
Average	T	Average	T
Positive composites		Positive composites	
Total number	15	Total number	5
Number reported as trace	7	Number reported as trace	4
Range	0.001–0.003	Range	0.0005
HEPTACHLOR EPOXIDE		LINDANE	
Average	T	Average	T
Positive composites		Positive composites	
Total number	13	Total number	2
Number reported as trace	10	Number reported as trace	2
Range	0.001	Range	T
SELENIUM		CADMIUM	
Average	0.004	Average	0.002
Positive composites		Positive composites	
Total number	4	Total number	3
Number reported as trace	0	Number reported as trace	0
Range	0.02–0.03	Range	0.01–0.02
OCTACHLOR EPOXIDE		ARSENIC	
Average	T	Average	0.004
Positive composites		Positive composites	
Total number	5	Total number	1
Number reported as trace	5	Number reported as trace	0
Range	T	Range	0.08
PCB			
Average	T		
Positive composites			
Total number	1		
Number reported as trace	1		
Range	T		
II. MEAT, FISH, AND POULTRY			
ZINC		SELENIUM	
Average	32.2	Average	0.20
Positive composites		Positive composites	
Total number	20	Total number	20
Number reported as trace	0	Number reported as trace	0
Range	25.3–76.0	Range	0.09–0.30
MERCURY		LEAD	
Average	0.02	Average	0.014
Positive composites		Positive composites	
Total number	18	Total number	5
Number reported as trace	0	Number reported as trace	0
Range	0.007–0.08	Range	0.05–0.07
CADMIUM		ARSENIC	
Average	0.01	Average	0.19
Positive composites		Positive composites	
Total number	17	Total number	17
Number reported as trace	0	Number reported as trace	0
Range	0.01–0.03	Range	0.03–0.46
p,p'-DDE		*p,p'*-TDE	
Average	0.010	Average	T
Positive composites		Positive composites	
Total number	20	Total number	7
Number reported as trace	1	Number reported as trace	6
Range	0.002–0.048	Range	0.004

TABLE 4. (cont'd.). *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

CHEMICAL	RESIDUES, PPM	CHEMICAL	RESIDUES, PPM
p,p'-DDT		DIELDRIN	
Average	0.002	Average	0.007
Positive composites		Positive composites	
Total number	15	Total number	19
Number reported as trace	9	Number reported as trace	1
Range	0.003–0.01	Range	0.001–0.086
α-BHC		HCB	
Average	T	Average	T
Positive composites		Positive composites	
Total number	19	Total number	11
Number reported as trace	17	Number reported as trace	8
Range	0.001	Range	0.0002–0.002
OCTACHLOR EPOXIDE		HEPTACHLOR EPOXIDE	
Average	T	Average	T
Positive composites		Positive composites	
Total number	12	Total number	15
Number reported as trace	10	Number reported as trace	9
Range	0.002	Range	0.001–0.002
RONNEL		PCB	
Average	T	Average	T
Positive composites		Positive composites	
Total number	1	Total number	2
Number reported as trace	0	Number reported as trace	2
Range	0.006	Range	T
LINDANE		*trans*-NONACHLOR	T
Average	T	Average	
Positive composites		Positive composites	
Total number	7	Total number	1
Number reported as trace	3	Number reported as trace	0
Range	0.0006–0.003	Range	0.002
ETHION			
Average	T		
Positive composites			
Total number	1		
Number reported as trace	1		
Range	T		

III. GRAIN AND CEREAL PRODUCTS

ZINC		SELENIUM	
Average	9.0	Average	0.19
Positive composites		Positive composites	
Total number	20	Total number	20
Number reported as trace	0	Number reported as trace	0
Range	5.5–15.5	Range	0.04–0.34
LEAD		CADMIUM	
Average	0.05	Average	0.03
Positive composites		Positive composites	
Total number	12	Total number	20
Number reported as trace	0	Number reported as trace	0
Range	0.04–0.14	Range	0.02–0.05
MALATHION		DIELDRIN	
Average	0.02	Average	T
Positive composites		Positive composites	
Total number	19	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.004–0.096	Range	0.003
CHLORDANE		ARSENIC	
Average	T	Average	0.02
Positive composites		Positive composites	
Total number	1	Total number	8
Number reported as trace	1	Number reported as trace	0
Range	T	Range	0.0303–0.10
MERCURY		DIAZINON	
Average	T	Average	T
Positive composites		Positive composites	
Total number	1	Total number	5
Number reported as trace	0	Number reported as trace	0
Range	0.01	Range	0.001–0.004

TABLE 4. (cont'd.). *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

CHEMICAL	RESIDUES, PPM	CHEMICAL	RESIDUES, PPM
TCNB		LINDANE	
Average	T	Average	T
Positive composites		Positive composites	
Total number	1	Total number	1
Number reported as trace	0	Number reported as trace	1
Range	0.002	Range	T
RONNEL			
Average	T		
Positive composites			
Total number	1		
Number reported as trace	0		
Range	0.002		

IV. POTATOES

ZINC		CADMIUM	
Average	5.18	Average	0.05
Positive composites		Positive composites	
Total number	20	Total number	20
Number reported as trace	0	Number reported as trace	0
Range	2.6–14.5	Range	0.02–0.09
TCNB		LEAD	
Average	0.007	Average	0.03
Positive composites		Positive composites	
Total number	6	Total number	5
Number reported as trace	0	Number reported as trace	0
Range	0.002–0.114	Range	0.06–0.32
HEPTACHLOR EPOXIDE		DIELDRIN	
Average	T	Average	T
Positive composites		Positive composites	
Total number	2	Total number	4
Number reported as trace	0	Number reported as trace	2
Range	0.001–0.002	Range	0.001–0.002
ENDOSULFAN SULFATE		SELENIUM	
Average	T	Average	0.006
Positive composites		Positive composites	
Total number	2	Total number	3
Number reported as trace	0	Number reported as trace	0
Range	0.003–0.012	Range	0.02–0.05
CIPC		CHLORDANE	
Average	0.04	Average	T
Positive composites		Positive composites	
Total number	5	Total number	1
Number reported as trace	0	Number reported as trace	1
Range	0.01–0.23	Range	T
p,p'-DDE			
Average	T		
Positive composites			
Total number	3		
Number reported as trace	2		
Range	0.003		

V. LEAFY VEGETABLES

ZINC		CADMIUM	
Average	2.67	Average	0.04
Positive composites		Positive composites	
Total number	20	Total number	19
Number reported as trace	0	Number reported as trace	0
Range	1.7–7.0	Range	0.02–0.10
ENDOSULFAN I		ENDOSULFAN II	
Average	0.001	Average	T
Positive composites		Positive composites	
Total number	5	Total number	5
Number reported as trace	0	Number reported as trace	2
Range	0.001–0.011	Range	0.002–0.004
ENDOSULFAN SULFATE		LINDANE	
Average	0.004	Average	T
Positive composites		Positive composites	
Total number	6	Total number	1
Positive composites	2	Number reported as trace	1
Range	0.008–0.030	Range	T

TABLE 4. (cont'd.). *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

CHEMICAL	RESIDUES, PPM	CHEMICAL	RESIDUES, PPM
PARATHION		**DIAZINON**	
Average	T	Average	T
Positive Composites		Positive composites	
Total number	1	Total number	2
Number reported as trace	0	Number reported as trace	1
Range	0.004	Range	0.003
TOXAPHENE		***p,p'*-DDE**	
Average	T	Average	0.003
Positive composites		Positive composites	
Total number	1	Total number	8
Number reported as trace	1	Number reported as trace	3
Range	T	Range	0.001–0.023
LEAD		***p,p'*-DDT**	
Average	0.007	Average	T
Positive composites		Positive composites	
Total number	2	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.06–0.09	Range	0.005
DICHLORAN		**DIELDRIN**	
Average	0.002	Average	T
Positive composites		Positive composites	
Total number	4	Total number	2
Number reported as trace	1	Number reported as trace	1
Range	0.002–0.025	Range	0.002
PERTHANE		**SELENIUM**	
Average	0.003	Average	T
Positive composites		Positive composites	
Total number	2	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.024–0.044	Range	0.010
DCPA		**β–BHC**	
Average	0.001	Average	T
Positive composites		Positive composites	
Total number	3	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.002–0.014	Range	0.008

VI. LEGUME VEGETABLES

ZINC		**LEAD**	
Average	7.62	Average	0.26
Positive composites		Positive composites	
Total number	20	Total number	17
Number reported as trace	0	Number reported as trace	0
Range	5.30–12.0	Range	0.08–0.82
CADMIUM		**ARSENIC**	
Average	0.01	Average	0.004
Positive composites		Positive composites	
Total number	14	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.01–0.07	Range	0.07
SELENIUM		**PARATHION**	
Average	0.008	Average	T
Positive composites		Positive composites	
Total number	5	Total number	2
Number reported as trace	0	Number reported as trace	0
Range	0.02–0.05	Range	0.002–0.003
***p,p'*-DDE**		**DICHLORAN**	
Average	T	Average	0.001
Positive composites		Positive composites	
Total number	1	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.001	Range	0.027

VII. ROOT VEGETABLES

ZINC		**LEAD**	
Average	2.32	Average	0.036
Positive composites		Positive composites	
Total number	20	Total number	8
Number reported as trace	0	Number reported as trace	0
Range	1.30–4.60	Range	0.06–0.14

TABLE 4. (cont'd.). *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

Chemical	Residues, ppm	Chemical	Residues, ppm
CADMIUM		**ARSENIC**	
Average	0.027	Average	0.004
Positive composites		Positive composites	
Total number	19	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.01–0.08	Range	0.070
SELENIUM		**PARATHION**	
Average	0.002	Average	T
Positive composites		Positive composites	
Total number	2	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.020	Range	0.005
DIELDRIN		***p,p'*–DDE**	
Average	T	Average	T
Positive composites		Positive composites	
Total number	1	Total number	3
Number reported as trace	1	Number reported as trace	1
Range	T	Range	0.004–0.009
DCPA		**ETHION**	
Average	T	Average	T
Positive composites		Positive composites	
Total number	2	Total number	1
Number reported as trace	1	Number reported as trace	1
Range	0.004	Range	T

VIII. GARDEN FRUITS

ZINC		**LEAD**	
Average	2.08	Average	0.081
Positive composites		Positive composites	
Total number	20	Total number	14
Number reported as trace	0	Number reported as trace	0
Range	1.20–3.50	Range	0.06–0.20
CADMIUM		**ARSENIC**	
Average	0.02	Average	0.005
Positive composites		Positive composites	
Total number	18	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.01–0.04	Range	0.10
DIELDRIN		**LEPTOPHOS**	
Average	0.002	Average	T
Positive composites		Positive composites	
Total number	13	Total number	2
Number reported as trace	2	Number reported as trace	1
Range	0.002–0.009	Range	0.009
ENDOSULFAN I		**ENDOSULFAN II**	
Average	T	Average	T
Positive composites		Positive composites	
Total number	6	Total number	4
Number reported as trace	1	Number reported as trace	2
Range	0.002–0.004	Range	0.004–0.005
ENDOSULFAN SULFATE		**CARBARYL**	
Average	T	Average	0.002
Positive composites		Positive composites	
Total number	3	Total number	2
Number reported as trace	2	Number reported as trace	1
Range	0.005	Range	0.050
DIAZINON		**LINDANE**	
Average	T	Average	T
Positive composites		Positive composites	
Total number	2	Total number	5
Number reported as trace	1	Number reported as trace	0
Range	0.002	Range	0.001–0.004
***p,p'*–DDE**		**PARATHION**	
Average	T	Average	T
Positive composites		Positive composites	
Total number	2	Total number	1
Number reported as trace	2	Number reported as trace	0
Range	T	Range	0.006
α–BHC		**ETHION**	
Average	T	Average	0.002
Positive composites		Positive composites	
Total number	3	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.004–0.007	Range	0.05

TABLE 4. (cont'd.). *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

Chemical	Residues, ppm	Chemical	Residues, ppm
CARBOFENOTHION		TCNB	
Average	0.010	Average	T
Positive composites		Positive composites	
Total number	1	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.195	Range	0.002
IX. FRUITS			
ZINC		LEAD	
Average	2.44	Average	0.041
Positive composites		Positive composites	
Total number	20	Total number	11
Number reported as trace	0	Number reported as trace	0
Range	0.10–19.0	Range	0.05–0.11
DICHLORAN		CARBARYL	
Average	0.009	Average	T
Positive composites		Positive composites	
Total number	4	Total number	3
Number reported as trace	0	Number reported as trace	3
Range	0.006–0.163	Range	T
PERTHANE		DICOFOL	
Average	T	Average	0.003
Positive composites		Positive composites	4
Total number	1	Number reported as trace	0
Number reported as trace	0	Range	0.007–0.028
Range	0.014		
ENDOSULFAN I		ENDOSULFAN II	
Average	T	Average	T
Positive composites		Positive composites	
Total number	1	Total number	2
Number reported as trace	0	Number reported as trace	1
Range	0.007	Range	0.012
ENDOSULFAN SULFATE		CADMIUM	
Average	T	Average	0.003
Positive composites		Positive composites	
Total number	2	Total number	5
Numuber reported as trace	1	Number reported as trace	0
Range	0.005	Range	0.01–0.02
PHOSALONE		ETHION	
Average	T	Average	T
Positive composites		Positive composites	
Total number	1	Total number	3
Number reported as trace	0	Number reported as trace	0
Range	0.008	Range	0.005–0.006
DIELDRIN		DIAZINON	
Average	T	Average	T
Positive composites		Positive composites	
Total number	2	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.001	Range	0.004
α-BHC		CAPTAN	
Average	T	Average	0.003
Positive composites		Positive composites	
Total number	1	Total number	2
Number reported as trace	0	Number reported as trace	0
Range	0.001	Range	0.026–0.040
MERCURY			
Average	T		
Positive composites			
Total number	1		
Number reported as trace	0		
Range	0.015		
X. OILS, FATS, AND SHORTENING			
ZINC		CADMIUM	
Average	4.14	Average	0.016
Positive composites		Positive composites	
Total number	20	Total number	18
Number reported as trace	0	Number reported as trace	0
Range	0.20–6.20	Range	0.01–0.03

TABLE 4. (cont'd.). *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

Chemical	Residues, ppm
MALATHION	
Average	0.003
Positive composites	
Total number	7
Number reported as trace	2
Range	0.005–0.03
DIELDRIN	
Average	T
Positive composites	
Total number	3
Number reported as trace	2
Range	0.002
ARSENIC	
Average	0.002
Positive composites	
Total number	1
Number reported as trace	0
Range	0.04
PCNB	
Average	T
Positive composites	
Total number	5
Number reported as trace	2
Range	0.002–0.003
α-BHC	
Average	T
Positive composites	
Total number	1
Number reported as trace	1
Range	T
LEAD	
Average	0.028
Positive composites	
Total number	6
Number reported as trace	0
Range	0.05–0.14
PENTACHLOROBENZENE	
Average	T
Positive composites	
Total number	2
Number reported as trace	0
Range	0.004–0.005
PCP METHYL ETHER	
Average	T
Positive composites	
Total number	1
Number reported as trace	0
Range	0.002
p,p'-DDE	
Average	T
Positive composites	
Total number	1
Number reported as trace	1
Range	T
MERCURY	
Average	T
Positive composites	
Total number	1
Number reported as trace	0
Range	0.008
HCB	
Average	T
Positive composites	
Total number	3
Number reported as trace	0
Range	0.001–0.006
PENTACHLOROANILINE	
Average	0.002
Positive composites	
Total number	4
Number reported as trace	1
Range	0.007–0.028
LINDANE	
Average	T
Positive composites	
Total number	1
Number reported as trace	1
Range	T
SELENIUM	
Average	0.004
Positive composites	
Total number	2
Number reported as trace	0
Range	0.04–0.05
PCTA	
Average	T
Positive composites	
Total numuber	2
Number reported as trace	0
Range	0.002–0.005

XI. SUGAR AND ADJUNCTS

ZINC	
Average	2.95
Positive composites	
Total number	20
Number reported as trace	0
Range	0.10–16.0
CADMIUM	
Average	0.011
Positive composites	
Total number	14
Number reported as trace	0
Range	0.01–0.03
LINDANE	
Average	T
Positive composites	
Total number	7
Number reported as trace	3
Range	0.001–0.003
PCP	
Average	0.003
Positive composites	
Total number	1
Number reported as trace	0
Range	0.06
o-PHENYLPHENOL	
Average	T
Positive composites	
Total number	1
Number reported as trace	1
Range	T
α-BHC	
Average	T
Positive composites	
Total number	5
Number reported as trace	5
Range	T

TABLE 4. (cont'd.). *Levels of chemical and metal residues, by food class, in adult food composites from 20 United States cities—August 1975–July 1976*

Chemical	Residues, ppm	Chemical	Residues, ppm
MALATHION		LEAD	
Average	T	Average	0.015
Positive composites		Positive composites	
Total number	3	Total number	3
Number reported as trace	1	Number reported as trace	0
Range	0.005–0.008	Range	0.06–0.14
DICHLORAN		MERCURY	
Average	T	Average	T
Positive composites		Positive composites	
Total number	1	Total number	1
Number reported as trace	0	Number reported as trace	0
Range	0.005	Range	0.012
	XII. BEVERAGES		
ZINC		LEAD	
Average	0.46	Average	0.004
Positive composites		Positive composites	
Total number	19	Total number	2
Number reported as trace	0	Number reported as trace	0
Range	0.20–1.90	Range	0.04–0.05
ARSENIC		CADMIUM	
Average	0.008	Average	0.002
Positive composites		Positive composites	
Total number	1	Total number	3
Number reported as trace	0	Number reported as trace	0
Range	0.15	Range	0.01
PCP		MERCURY	
Average	0.001	Average	0.001
Positive composites		Positive composites	
Total number	1	Total number	2
Number reported as trace	0	Number reported as trace	0
Range	0.026	Range	0.006–0.018

NOTE: Average values are based on 20 composites examined; trace residues, if present, were treated as zero in calculating averages. Thus, an average value of "T" can be well below detection limits of the methods for that compound.

TABLE 5. *FY 76 daily intakes, by food group, of metals in the diet of United States adults*

Food Group	Lead		Cadmium		Zinc		Arsenic [1]		Selenium		Mercury	
	µg/day	% Total Intake	µg/day	% Total Intake	mg/day	% Total Intake	µg/day	% Total Intake	µg/day	% Total Intake	µg/day	% Total Intake
I. Dairy products	0.00	0.0	1.63	5.0	3.72	19.4	3.17	4.8	3.26	2.4	0.00	0.0
II. Meat, fish, and poultry	3.67	5.2	2.63	8.0	8.44	44.1	49.46	74.4	52.52	38.7	5.29	81.3
III. Grains and cereal products	19.90	28.0	11.97	36.4	3.81	19.9	7.56	11.4	78.41	57.8	0.21	3.2
IV. Potatoes	5.15	7.2	7.46	22.7	0.82	4.3	0.00	0.0	0.59	0.4	0.00	0.0
V. Leafy vegetables	0.38	0.5	2.43	7.4	0.15	0.8	0.00	0.0	0.03	0.02	0.00	0.0
VI. Legume vegetables	18.98	26.7	0.82	2.5	0.56	2.9	0.28	0.4	0.61	0.4	0.00	0.0
VII. Root vegetables	1.27	1.8	0.87	2.6	0.08	0.4	0.10	0.1	0.06	0.04	0.00	0.0
VIII. Garden fruits	6.07	8.5	1.42	4.3	0.15	0.8	0.36	0.5	0.00	0.0	0.00	0.0
IX. Fruits	9.07	12.8	0.66	2.0	0.54	2.8	0.00	0.0	0.00	0.0	0.10	1.5
X. Oils, fats, and shortening	2.04	2.9	1.11	3.4	0.30	1.6	0.00	0.0	0.16	0.1	0.02	0.3
XI. Sugar and adjuncts	1.24	1.7	0.87	2.6	0.24	1.3	0.00	0.0	0.00	0.0	0.04	0.6
XII. Beverages (including drinking water)	3.31	4.7	1.02	3.1	0.32	1.7	5.57	8.4	0.00	0.0	0.85	13.1
Total intake	71.08	100.0	32.89	100.0	19.13	100.0	66.50	100.0	135.64	99.9[2]	6.51	100.0

[1] Values calculated as arsenic trioxide (As_2O_3).
[2] Does not total 100 because of rounding error.

organochlorine compounds present at low levels included α-BHC (hexachlorocyclohexane), dieldrin, heptachlor epoxide, octachlor epoxide, methoxychlor, HCB (hexachlorobenzene), and lindane. A trace of an industrial chemical, a PCB (polychlorinated biphenyl), was found in one of the composites. No organophosphorus compounds were found.

MEAT, FISH, AND POULTRY

Metal residues dominated this food class, with the following series averages: 32.2 ppm zinc (range 25.3–76.0 ppm), 0.20 ppm selenium, 0.19 ppm arsenic, 0.02 ppm mercury, 0.014 ppm lead, and 0.01 ppm cadmium. Of the organochlorine residues, *p,p'*-DDE, ranging from 0.002 to 0.048 ppm and averaging 0.010 ppm for the

series, was found in all 20 composites; dieldrin, averaging 0.007 ppm, was reported in 19 composites; and *p,p'*-DDT averaged 0.002 ppm with positive findings in 15 composites. Trace averages were reported for *p,p'*-TDE, α-BHC, HCB, octachlor epoxide, heptachlor epoxide, ronnel, lindane, and *trans*-nonachlor. Traces of ethion, an organophosphorus pesticide, and a PCB also were found in one and two composites, respectively.

GRAIN AND CEREAL PRODUCTS

All 20 composites contained zinc, selenium, and cadmium residues, averaging 9.0, 0.19, and 0.03 ppm, respectively. Twelve composites had lead residues, averaging 0.05 ppm for the series, and eight composites had arsenic residues, for a series average of 0.02 ppm. Malathion, one of two organophosphorus pesticides, was reported in 19 composites and averaged 0.02 ppm for the series; the other, ronnel, occurred as a trace amount in one composite. Also reported were traces of dieldrin, chlordane, TCNB, lindane, diazinon, and mercury.

POTATOES

Zinc, ranging from 2.6 to 14.5 ppm, and cadmium, ranging from 0.02 to 0.09 ppm, were reported for all 20 composites, with averages of 5.18 and 0.05 ppm, respectively. Lead, averaging 0.03 ppm for the series, was found in five composites, and selenium, ranging from 0.02 to 0.05 ppm in three composites, averaged 0.006 ppm for the 20-composite series. CIPC, ranging from 0.01 to 0.23 ppm in five composites, averaged 0.04 ppm for the series. TCNB averaged 0.007 ppm for the series with a range of 0.002–0.114 ppm in six composites. Traces of heptachlor epoxide, dieldrin, endosulfan sulfate, *p,p'*-DDE, and chlordane were also found.

LEAFY VEGETABLES

Only zinc, ranging from 1.7 to 7.0 ppm and averaging 2.67 ppm, was reported for all 20 composites. Cadmium ranged from 0.02 to 0.10 ppm in 19 composites and averaged 0.04 ppm for the series. The most frequently reported pesticide was *p,p'*-DDE, averaging 0.003 ppm for the series, with eight reported findings. Endosulfan I, ranging from 0.001 to 0.011 ppm, and endosulfan II, ranging from 0.002 to 0.004 ppm, were each reported for five composites; endosulfan sulfate, ranging from 0.008 to 0.030 ppm, was reported for six composites. Other reportable residues and their averages included dichloran, 0.002 ppm; DCPA, 0.001 ppm; Perthane®, 0.003 ppm; and lead, 0.007 ppm. Traces of lindane, diazinon, parathion, toxaphene, dieldrin, *p,p'*-DDT, selenium, and β-BHC were also found.

LEGUME VEGETABLES

Legume vegetables exhibited high metal residues. Zinc, reported in all 20 composites, ranged from 5.30 to 12.0 ppm and averaged 7.62 ppm. Lead ranged from 0.08 to 0.82 ppm and averaged 0.26 ppm for the series. Cadmium, ranging from 0.01 to 0.07 ppm, averaged 0.01 ppm for the series. Arsenic and selenium occurred less frequently. The pesticides parathion, *p,p'*-DDE, and dichloran were found at low levels.

ROOT VEGETABLES

Zinc ranged from 1.30 to 4.60 ppm and averaged 2.32 ppm for the 20 composites. Cadmium, ranging from 0.01 to 0.08 ppm in 19 composites, averaged 0.027 ppm overall. Lesser amounts of lead, arsenic, and selenium, averaging 0.036, 0.004, and 0.002 ppm, respectively, were also reported. Only traces of parathion, dieldrin, *p,p'*-DDE, DCPA, and ethion were found.

GARDEN FRUITS

Four metals were reported in this food class: zinc, ranging from 1.20 to 3.50 ppm in 20 composites and averaging 2.08 ppm; cadmium, ranging from 0.01 to 0.04 ppm in 18 composites and averaging 0.02 ppm for the 20-composite series; lead, ranging from 0.06 to 0.20 ppm in 14 composites and averaging 0.081 ppm for the series; and arsenic, reported in one composite at 0.10 ppm. The most significant pesticide residue, dieldrin, ranged from 0.002 to 0.009 ppm in 13 composites and averaged 0.002 ppm for the series. Carbophenothion was found in one composite at 0.195 ppm, with a series average of 0.01 ppm. The following trace averages also were reported: leptophos; endosulfan I, II, and sulfate; carbaryl; diazinon; lindane; *p,p'*-DDE; parathion; α-BHC; ethion; and TCNB.

FRUITS

The two most prevalent residues in this food class were zinc, reported for all 20 composites, ranging from 0.10 to 19.0 ppm and averaging 2.44 ppm, and lead, ranging from 0.05 to 0.11 ppm in 11 composites and averaging 0.041 ppm for the series. Cadmium, ranging from 0.01 to 0.02 ppm in five composites, averaged 0.003 ppm for the series. Both dichloran and dicofol were found in four composites and averaged 0.009 ppm and 0.003 ppm, respectively, for the series. Ethion ranged from 0.005 to 0.006 ppm for three composites but averaged trace for the series. Less frequently occurring residues were carbaryl, averaging a trace for the series; captan; endosulfan I, II, and sulfate; Perthane; phosalone; diazinon; dieldrin; and α-BHC. Mercury, 0.015 ppm, was reported for one composite.

OILS, FATS, AND SHORTENING

High zinc levels, ranging from 0.20 to 6.20 ppm and averaging 4.14 ppm, were reported for 20 composites. Cadmium, ranging from 0.01 to 0.03 ppm in 18 composites, averaged 0.016 ppm for the series. Seven composites contained malathion residues, ranging from 0.005 to 0.03 ppm; series average was 0.003. Lead, averaging 0.028 ppm for the series, ranged from 0.05 to 0.14 ppm

TABLE 6. *Pesticide residues in individual commodities of dairy composite of four market basket samples—August 1975–July 1976*

Residue Found	Commodity [1,2]								
	Whole Milk (4)	Evaporated Milk (4)	Ice Cream (4)	Cottage Cheese (4)	Processed Cheese (4)	Natural Cheese (4)	Butter (4)	Skim Milk (4)	Ice Milk (2)
α-BHC									
Times found	1	4	4	3	4	4	4		2
Range, ppm	T	T–0.002	T–0.002	T	0.002–0.005	0.001–0.008	0.008–0.011		T–0.001
p,p'-DDE									
Times found	2	4	3	2	4	3	4		2
Range, ppm	0.002–0.003	T–0.020	0.002–0.010	0.003–0.009	0.002–0.016	T–0.004	0.004–0.132		0.002–0.006
HCB									
Times found	1	2	3	1	3	3	4	1	1
Range, ppm	T	T	T	0.001	T–0.001	T–0.002	T–0.004	T	T
Heptachlor epoxide									
Times found		2	2	1	3	4	4		
Range, ppm		T	T	T	0.002–0.005	T–0.004	0.004–0.019		
Dieldrin									
Times found		4	4	2	4	4	4		
Range, ppm		T–0.002	T–0.002	T	0.005–0.010	0.003–0.010	0.017–0.053		
Methoxychlor									
Times found		1	1	1		1	1		
Range, ppm		0.016	T	T		T	0.141		
Lindane									
Times found			2		1		2		
Range, ppm			T		0.001		T–0.002		
Octachlor epoxide									
Times found			1		3	3	3		
Range, ppm			T		T–0.002	T	0.004–0.008		
p,p'-DDT									
Times found					2	1			
Range, ppm					T	T			
p,p'-TDE									
Times found						1			
Range, ppm						T			

NOTE: T = trace.
[1] Buttermilk and nonfat dry milk not included because no residues were found
[2] Numbers in parentheses indicate number of times that commodity was analyzed.

in six composites. Pentachloroaniline, averaging 0.002 ppm for the series, was found in four composites. The remaining residues included HCB, selenium, PCNB, PCTA, pentachlorobenzene, dieldrin, α-BHC, arsenic, *p,p'*-DDE, and lindane. One composite contained 0.008 ppm mercury.

SUGAR AND ADJUNCTS

The metal residues were among the highest in this food class. Zinc, ranging from 0.10 to 16.0 ppm, was reported in all 20 composites and averaged 2.95 ppm. Cadmium, found in 14 composites, averaged 0.011 ppm for the series. Lead, ranging from 0.06 to 0.14 ppm in three composites, averaged 0.015 ppm for the series. Mercury at 0.012 ppm was reported for one composite. Other residues were α-BHC, PCP, lindane, malathion, *o*-phenylphenol, and dichloran.

BEVERAGES

Zinc, found in 19 composites at levels ranging from 0.20 to 1.90 ppm, averaged 0.46 ppm for the series. The remaining residues, each found in three or fewer composites, had the following series averages: cadmium, 0.002 ppm; lead, 0.004 ppm; mercury, 0.001 ppm; arsenic, 0.008 ppm; and PCP, 0.001 ppm.

Discussion

Of the 240 composites analyzed, 125, or 52 percent, contained organochlorine pesticide residues, compared with 49, 48, 52, and 54 percent reported for fiscal years 1975, 1974, 1973, and 1972, respectively. Organophosphorus residues in the current reporting period were found in 45, or 18.7 percent, of the composites. Corresponding findings for fiscal years 1975, 1974, 1973, and 1972 were 25, 28, 31, and 28 percent, respectively. The present report and that for FY 75 were based on 20 market baskets, whereas all earlier reports were based on 30 market baskets.

Sixty percent of the 346 organochlorine residues in the current reporting period were found in two food classes: dairy products and meat-fish-poultry. The remaining organochlorine residues were distributed among the other food classes with the garden fruits and leafy vegetables containing half of them. No organochlorine resi-

TABLE 7. *Pesticide residues in individual commodities of meat and fish composites of four market baskets, August 1975–July 1976*

Residue Found	Commodity [1]						
	Roast Beef (4)	Ground Beef (4)	Pork Chops (4)	Bacon (4)	Chicken (4)	Fish Fillet (4)	Tuna (4)
Dieldrin							
Times found	4	2	4	2	4		1
Range, ppm	T–0.003	0.006–0.007	0.002–0.006	T–2.25	0.002–0.007		T
p,p'-TDE							
Times found	1		1	1		2	4
Range, ppm	0.008		0.016	0.007		T–0.081	T–0.018
HCB							
Times found	3	2	1	1	2	2	1
Range, ppm	T–0.002	0.002	0.002	0.030	0.002–0.008	T–0.003	T
α-BHC							
Times found	2	2		1		2	1
Range, ppm	T–0.001	0.001		T		0.002	T
p,p'-DDE							
Times found	3	1	2	2	4	3	4
Range, ppm	T–0.048	0.031	0.002–0.152	0.002–0.019	0.002–0.028	0.010–0.875	T–0.015
Heptachlor epoxide							
Times found	2	2	2		2		
Range, ppm	0.001–0.009	0.002	T–0.005		T		
Ethion							
Times found		1					
Range, ppm		0.05					
Octachlor epoxide							
Times found	1	1	3	1			
Range, ppm	0.002	0.002	T–0.011	0.003			
trans-Nonachlor							
Times found			1	1			
Range, ppm			0.034	0.003			
p,p'-DDT							
Times found			1	2		3	
Range, ppm			0.034	T–0.141		0.023–0.130	
Endrin							
Times found					1		
Range, ppm					0.001		
PCB (Aroclor 1254)							
Times found	1				1	T	
Range, ppm	T				T	1	
Chlordane							
Times found					1	1	
Range, ppm					T	T	
Lindane							
Times found							
Range, ppm							
TCNB							
Times found							
Range, ppm							

NOTE: T = trace.
[1] Numbers in parentheses indicate the number of times the item was analyzed.

Commodity[1]								
Luncheon Meat (4)	Frankfurters (4)	Beef Liver (4)	Eggs (4)	Ham (4)	Round Steak (4)	Veal (1)	Lamb (2)	Shrimp (2)
4 0.002–0.006	4 0.004–0.054	1 0.014	3 T–0.018	2 T–0.002	2 0.002–0.004	1 T	1 T	
2 T–0.002	2 0.001–0.002	1 T	2 T		2 T		2 0.002	
4 0.001–0.003	4 T–0.004	3 T–0.004			3 T		2 T–0.003	
4 0.004–0.030	4 T–0.029	2 T–0.003	3 0.002–0.026	1 0.002	2 0.006–0.026	1 T	2 0.011–0.037	1 T
3 0.001–0.002	3 T–0.002	1 0.005			1 0.002			
4 T–0.003	2 T–0.002	1 T					1 0.002	
1 0.005								
2 T–0.011								
			1 T					
2 T–0.002	1 0.003							
		1 T						

TABLE 8. *Recovery data on residues found in adult total diet samples, August 1975–July 1976*

Residue	Type of Food Composite	Spike Level, ppm	Range of Unfortified Composite, ppm	Range of Total Residue Found, ppm [1,2]	No. of Recovery Attempts
Heptachlor epoxide	fatty	0.003	0–0.001	0.0023–0.0035 (0.0031)	4
	nonfatty	0.003	0–0.001	0.0027–0.0039 (0.0032)	6
Oxychlordane	fatty	0.003	0–0.0007	0.0022–0.0031 (0.0027)	4
	nonfatty	0.003	0–0.0006	0.0024–0.0035 (0.0027)	6
Ethion	fatty	0.01	0.00	0–0.0056 (—)	4
	nonfatty	0.01	0.00	0.0066–0.012 (0.0086)	6
DCPA	fatty	0.005	0.00	0.0027–0.0047 (0.0037)	6
	nonfatty	0.005	0–0.0084	0.0029–0.0059 (0.0050)	12
Perthane	fatty	0.01	0.00	T–0.0104 (0.0069)	6
	nonfatty	0.01	0.00	0.0035–0.014 (0.0091)	12
Methyl parathion	fatty	0.005	0.00	0.0022–0.0030 (0.0026)	3
	nonfatty	0.005	0.00	0.0031–0.0045 (0.0037)	5
Endosulfan sulfate	fatty	0.01	0.00	0.002–0.0076 (0.0048)	2
	nonfatty	0.01	0.00–0.004	0.0035–0.0143 (0.0095)	6
Tetradifon	fatty	0.02	0.00	T–0.027 (0.015)	4
		0.10	0.00	0.088–0.089 (—)	2
	nonfatty	0.02	0.00	0.0104–0.022 (0.0160)	8
		0.10	0.00	0.054–0.188 (0.109)	6
Malathion	fatty	0.005	0.00	0.0037–0.0038 (—)	2
	nonfatty	0.005	0.00	0.0022–0.0064 (0.0041)	6
Phosalone	fatty	0.02	0.00	0.0–0.017 (—)	3
	nonfatty	0.02	0.00–0.002	0.007–0.026 (0.0166)	6
Leptophos	fatty	0.05	0.00	0.016–0.040 (0.027)	3
	nonfatty	0.05	0.00	0.030–0.053 (0.043)	6
Fonofos	fatty	0.01	0.00	0.002	1
	nonfatty	0.01	0.00	0.0067–0.0095 (0.0083)	6
Toxaphene	fatty	0.20	0.00	0.148	1
	nonfatty	0.20	0.00	0.147–0.226 (0.187)	6
2,4-D	fatty	0.04	0.00–0.009	0.00–0.050 (0.021)	7
	nonfatty	0.04	0.00	0.008–0.046 (0.028)	14
PCP	fatty	0.02	0.00–0.003	0.0012–0.009 (0.007)	4
		0.04	0.00–0.001	0.00–0.030 (0.015)	7
	nonfatty	0.02	0.0–0.0004	0.003–0.018 (0.0084)	1
		0.04	0.00–0.0062	0.003–0.039 (0.022)	1
Picloran	fatty	0.10	0.00	0.00–0.043 (—)	2
	nonfatty	0.10	0.00	0.033–0.075 (0.046)	4
Silvex	fatty	0.04	0.00	0.00–0.032 (0.015)	5
	nonfatty	0.04	0.00	0.020–0.053 (0.033)	9
2,4-DB	fatty	0.02	0.00	T–0.005 (—)	2
		0.04	0.00	0.00–0.033 (0.014)	4
	nonfatty	0.02	0.00	0.002–0.015 (0.008)	4
		0.04	0.00	0.00–0.038 (0.025)	8
2,4,5-T	fatty	0.02	0.00–0.001	0.0058–0.021 (—)	2
		0.08	0.00	0.026–0.059 (—)	2
	nonfatty	0.02	0.00	0.003–0.0198 (0.014)	4
		0.08	0.00	0.032–0.069 (0.048)	4
MCP 2-methyl-4-chlorophenoxy-acetic acid	fatty	0.02	0.00	0.009–0.013 (0.011)	3
	nonfatty	0.02	0.00	0.0055–0.026 (0.014)	5
Carbaryl	nonfatty	0.20	0.00	0.00–0.20 (0.157)	39
o-Phenylphenol	nonfatty	0.40	0.00	0.00–0.40 (0.24)	37
Arsenic	fatty	0.30	0.00–0.35	0.26–0.81 (0.376)	5
		0.40	0.00–0.28	0.37–0.86 (0.44)	10
	nonfatty	0.30	0.00–0.03	0.19–0.46 (0.30)	10
		0.40	0.00–0.03	0.13–0.57 (0.44)	21
Cadmium	fatty	0.10	0.00–0.027	0.094–0.199 (0.113)	21
	nonfatty	0.10	0.00–0.058	0.058–0.156 (0.116)	38
Lead	fatty	0.20	0.00–0.14	0.020–0.340 (0.133)	18
	nonfatty	0.20	0.00–0.310	0.058–0.480 (0.211)	34
Mercury	fatty	0.06	0.00–0.049	0.018–0.110 (0.073)	15
	nonfatty	0.06	0.00–0.011	0.062–0.088 (0.075)	26
Selenium	fatty	0.20	0.00–0.23	0.00–0.39 (0.227)	12
	nonfatty	0.20	0.00–0.24	0.11–0.48 (0.216)	28
Zinc	fatty	5.0	1.23–6.20	5.74–11.00 (9.54)	11
		25.0	4.00–76.00	29.00–99.00 (54.2)	8
	nonfatty	5.0	0.10–12.00	4.68–15.0 (8.09)	26
		25.0	0.38–15.50	24.38–38.0 (29.8)	14

NOTE: T = trace.
[1] Numbers in parentheses represent average residue levels.
[2] These values are uncorrected for background.

dues were found in beverage composites and only two were found in the legume vegetable composite.

The 56 organophosphorus residues constitute about 14 percent of the total pesticide residues reported, with malathion representing 29 of them. Nineteen were found in grain and cereal products, seven in fats and oils, and three in the sugar composites. No organophosphorus residues were found in the dairy products, potatoes, or beverage composites.

The carbamate pesticide carbaryl occurred in five composites, once at the 0.05 ppm level and four times at the trace level. The method for determination of carbaryl will also detect the fungicide *o*-phenylphenol, which was reported in one sugar composite at the trace level.

Two industrial chemicals were detected. Trace amounts of a PCB, Aroclor 1254, were found in one dairy composite and in two meat-fish-poultry composites. Low levels of pentachlorobenzene were found in two fat composites.

Of all the residues reported, 606, or 58 percent, were metals. Zinc was reported in almost every composite, with the highest levels being found in the meat composites. Cadmium was found in 170 composites and lead in 85 composites; both were found throughout the various classes of foods with fewest findings in the dairy and the beverage composites. The 57 selenium residues, 31 arsenic residues, and 24 mercury residues were found predominantly in the meat-fish-poultry composites and grain-cereal composites.

In addition to the analysis of the various food class composites, four market baskets, one from each region, were selected for individual item analysis of two food groups. Such item-by-item analysis often provides a more explicit picture as to the source of residues within a composite. The dairy and meat classes were chosen because those composites have consistently contained the highest levels of organochlorine and organophosphorus residues. Tables 6 and 7 present these findings.

Recovery studies were conducted with each market basket. Composites were fortified with known compounds representing each type of residue (metal, pesticide, etc.), and corrections were made for the unfortified composite contribution. The total amount recovered through the method was determined. These results are presented in Table 8.

Acknowledgments

The authors acknowledge the contributions of all staff members assigned to the Total Diet Section, Food and Drug Administration, Kansas City District Laboratory.

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APPENDIX

Chemical Names of Compounds Discussed in This Issue

ALDRIN	Hexachlorohexahydro-*endo,exo*-dimethanonaphthalene 95% and related compounds 5%
AROCLOR 1016 or 1242	PCB, approximately 42% chlorine
AROCLOR 1242	PCB, approximately 42% chlorine
AROCLOR 1254	PCB, approximately 54% chlorine
BHC (Benzene Hexachloride)	1,2,3,4,5,6-Hexachlorocyclohexane (mixture of isomers)
CAPTAN	*N*-Trichloromethylthio-4-cyclohexene-1,2-dicarboximide
CARBARYL	1-Naphthyl methylcarbamate
CARBOPHENOTHION	*S*-[[(*p*-Chlorophenyl)thio]methyl] *O,O*-diethyl phosphorodithioate
CHLORDANE	Technical: 60% octachloro-4,7-methanotetrahydroindane and 40% related compounds
CIPC	Isopropyl *N*-(3-chlorophenyl)carbamate
2,4-D	2,4-Dichlorophenoxyacetic acid
DACTHAL	Dimethyl tetrachloroterephthalate
2,4-DB	4-(2,4-Dichlorophenoxy)butyric acid
DDE	Dichlorodiphenyldichloroethylene (degradation product of DDT)
DDMU	1-Chloro-2,2-bis(*p*-chlorophenyl) ethylene
DDT	Dichloro diphenyl trichloroethane. Principal isomer present (*p,p'*-DDT; not less than 70%: 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane
DIAZINON	*O,O*-Diethyl *O*-(2-isopropyl-6-methyl-4-pyrimidinyl) phosphorothioate
DICHLORAN	2,6-Dichloro-4-nitroaniline
DICOFOL	1,1-Bis(chlorophenyl)-2,2,2-trichloroethanol
DIELDRIN	Hexachloroepoxyoctahydro-*endo,exo*-dimethanonaphthalene 85% and related compounds 15%
ENDOSULFAN	Hexachlorohexahydromethano-2,4,3-benzodioxathiepin 3-oxide
ENDOSULFAN SULFATE	1,4,5,6,7,7-Hexachloro-5-norbornene-2,3-dimethanol cyclic sulfate
ENDRIN	Hexachloroepoxyoctahydro-*endo,endo*-dimethanonaphthalene
ETHION	*O,O,O',O'*-Tetraethyl *S,S'*-methylene bisphosphorodithioate
FONOFOS	*O*-Ethyl *S*-phenyl ethylphosphonodithioate
HCB	Hexachlorobenzene
HEPTACHLOR	Heptachlorotetrahydro-4,7-methanoindene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-Heptachloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindan
LEPTOPHOS	*O*-(4-Bromo-2,5-dichlorophenyl) *O*-methyl phenylphosphonothioate
LINDANE	*Gamma* isomer of benzene hexachloride (BHC)
MALATHION	*O,O*-Dimethyl dithiophosphate of diethyl mercaptosuccinate
MCP	See MCPA

(*Continued next page*)

APPENDIX (*continued*)

MCPA	2-Methyl-4-chlorophenoxyacetic acid
METHOXYCHLOR	2,2-Bis(*p*-methoxyphenyl),1,1,1-trichloroethane 88% and related compounds 12%
METHYL PARATHION	*O,O*-Dimethyl *O*-*p*-nitrophenyl phosphorothioate
MIREX	Dodecachlorooctahydro-1,3,4-metheno-1*H*-cyclobuta[cd]pentalene
NONACHLOR	1,2,3,4,5,6,7,8,8-Nonachloro-3a,4,7,7a-tetrahydro-4,7-methanoindan
OCTACHLOR EPOXIDE	1-*exo*-2-*endo*-4,5,6,7,8,8a-Octachloro-2,3-*exo*-epoxy-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene
OXYCHLORDANE	1-*exo*-2-*endo*-4,5,6,7,8,8a-Octachloro-2,3-*exo*-epoxy-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene
PARATHION	*O,O*-Diethyl *O*-*p*-nitrophenyl phosphorothioate
PCA	Pentachloroaniline
PCBs (Polychlorinated Biphenyls)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorine
PCNB	Pentachloronitrobenzene
PCP	Pentachlorophenol
PCTA	Pentachlorothioanisole
PCTs (Polychlorinated Terphenyls)	Mixtures of chlorinated terphenyl compounds having various percentages of chlorine
PERTHANE	1,1-Bis(ethylphenyl)-2,2-dichloroethane
PHOSALONE	*S*-[6-Chloro-3-(mercaptomethyl)-2-benzoxazolinone] *O,O*-diethyl phosphorodithioate
PHOTOMIREX	1,2,3,4,5,5,6,7,9,10,10-Undecachloropentacyclo[5.3.0.$0^{2,6}$.$0^{3,9}$.$0^{4,8}$]decane
PICLORAM	4-Amino-3,5,6-trichloropicolinic acid
RONNEL	*O,O*-Dimethyl *O*-(2,4,5-trichlorophenyl) phosphorothioate
2,4,5-T	2,4,5-Trichlorophenoxyacetic acid
TCNB	1,2,4,5-Tetrachloro-3-nitrobenzene
TCTA	Tetrachlorothioanisole
TDE	Dichloro diphenyl dichloroethane (1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane, principal component)
TETRADIFON	4-Chlorophenyl 2,4,5-trichlorophenyl sulfone
TOXAPHENE	Technical chlorinated camphene (67-69% chlorine)
2,4,5-TP	2-(2,4,5-Trichlorophenoxy)propionic acid

ERRATUM

Pesticides Monitoring Journal, Volume 14, Number 4, page 136. The abstract of the article "Organochlorine Residues in Fish: National Pesticide Monitoring Program, 1970-74" should be corrected to read as follows:

Highest PCB residues were found in the industrialized areas of the Northeast and Midwest. . . .

PLANT GROWTH REGULATOR WORKING GROUP
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The Plant Growth Regulator Working Group (PGRWG) is announcing its eighth annual meeting, to be held at the Don Cesar Hotel, St. Petersburg, Florida, August 3–6, 1981.

The meeting will feature two symposia:

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Fifty-two technical papers will be presented by industry and university scientists, covering a range of chemicals being tested or having potential for future food production and crop management.

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The *Pesticides Monitoring Journal* welcomes from all sources qualified data and interpretative information on pesticide monitoring. The publication is distributed principally to scientists, technicians, and administrators associated with pesticide monitoring, research, and other programs concerned with pesticides in the environment. Other subscribers work in agriculture, chemical manufacturing, food processing, medicine, public health, and conservation.

Articles are grouped under seven headings. Five follow the basic environmental components of the National Pesticide Monitoring Program: Pesticide Residues in People; Pesticide Residues in Water; Pesticide Residues in Soil; Pesticide Residues in Food and Feed; and Pesticide Residues in Fish, Wildlife, and Estuaries. The sixth is a general heading; the seventh encompasses briefs.

Monitoring is defined here as the repeated sampling and analysis of environmental components to obtain reliable estimates of levels of pesticide residues and related compounds in these components and the changes in these levels with time. It can include the recording of residues at a given time and place, or the comparison of residues in different geographic areas. The Journal will publish results of such investigations and data on levels of pesticide residues in all portions of the environment in sufficient detail to permit interpretations and conclusions by author and reader alike. Such investigations should be specifically designed and planned for monitoring purposes. The Journal does not generally publish original research investigations on subjects such as pesticide analytical methods, pesticide metabolism, or field trials (studies in which pesticides are experimentally applied to a plot or field and pesticide residue depletion rates and movement within the treated plot or field are observed).

Authors are responsible for the accuracy and validity of their data and interpretations, including tables, charts, and references. Pesticides ordinarily should be identified by common or generic names approved by national or international scientific societies. Trade names are acceptable for compounds which have no common names. Structural chemical formulas should be used when appropriate. Accuracy, reliability, and limitations of sampling and analytical methods employed must be described thoroughly, indicating procedures and controls used, such as recovery experiments at appropriate levels, confirmatory tests, and application of internal standards and interlaboratory checks. The procedure employed should be described in detail. If reference is made to procedures in another paper, crucial points or modifications should be noted. Sensitivity of the method and limits of detection should be given, particularly when very low levels of pesticide residues are being reported. Specific note should be made regarding correction of data for percent recoveries. Numerical data, plot dimensions, and instrument measurements should be reported in metric units.

PREPARATION OF MANUSCRIPTS

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